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Superstructures in Alloy Systems

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THE existence of superstructures in alloy systems was postulated by Tammann[1] in 1919. He observed that when a series of well-annealed binary alloys, containing one noble component, are exposed to a corrosive agent (which attacks the base metal component but not the noble one) a critical composition exists such that corrosion is rapid in alloys containing less of the noble component than corresponds to this critical composition, whereas in alloys richer in the noble metal no corrosion is detectable. The locations of these so-called resistance limits were found to be functions of the nature and concentration of the corrosive agent, but they invariably occurred near compositions of either 25 or 50 atomic percent of the noble metal. These facts led Tammann to postulate the existence of what he called normal structure or as we know it today, a superstructure, which is the equilibrium structure attained in certain composition regions on slow cooling, or by prolonged annealing at low temperatures which are yet sufficiently high to permit diffusion to take place.

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Although additional x-ray lines, over those characteristic of a face-centered cubic lattice, were observed by Bain[2] in diffraction patterns obtained from a Cu_3Au alloy which had been slowly cooled, it was not until 1925 that Johansson and Linde[3] were able to obtain definite x-ray evidence for the existence of a superlattice in the Cu-Au system. These superstructure lines arise by virtue of the difference in scattering power for x-rays of the different kinds of atoms arranged in an orderly array in the phase sites and are only detectable if this difference is sufficiently great. The Cu-Au system is one which lends itself to careful structure study by means of x-rays.

Johansson and Linde found that a copper-gold alloy containing 50 atomic percent gold, when rapidly cooled or "quenched" from a temperature above 450°C so as to retain the condition prevailing at the elevated temperature, is a solid

solution with the atoms of Cu and Au statistically distributed among the lattice sites of the face-centered lattice. The same alloy, after slow cooling from temperatures above 430°C or after annealing at *ca.* 300°C over the several hours required for the alloy to relax into the equilibrium state, possesses a so-called superlattice structure: The lattice suffers a change from face-centered cubic to tetragonal. The atoms of copper occupy the tetragonal prism corners and basal face-centers while the gold atoms occupy the lateral face-centers. Figs. 1A and C, show the distribution of the atoms among the phase sites for both the random and ordered structures of an alloy containing 50 atomic percent gold. The ordered structure there depicted is that which should prevail under conditions permitting perfect ordering. Johansson and Linde[4] have recently shown that the annealing of Cu-Au alloys containing from 36 to 47 and from 53 to 65 atomic percent gold at temperatures between 200 and 400°C, or the quenching of alloys containing 47 to 53 atomic percent from temperatures between 410°–420°C, produces an ordered structure with orthorhombic symmetry.

Figure 1B depicts the condition of perfect order in a Cu_3Au alloy. The full circles represent the gold atoms on the cube corners and the empty circles the copper atoms at face-centers.

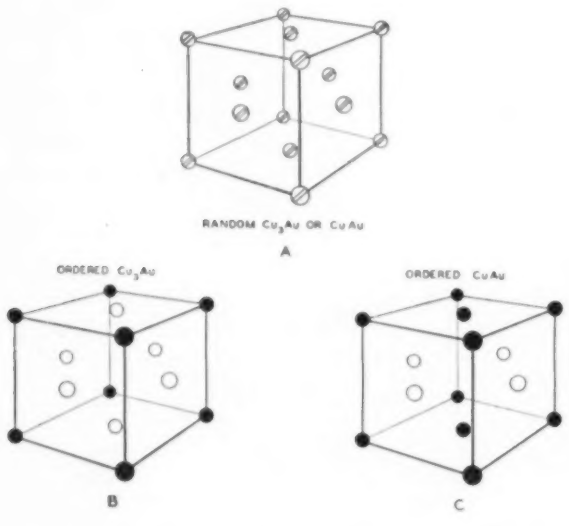

FIG. 1. Distribution of atoms; (*A*) random face-centered cubic of Cu_3Au or CuAu; (*B*) ordered cubic Cu_3Au (*C*) ordered tetragonal CuAu. Cu, empty circles; Au, full circles.

Similar structures have been observed[5] in Cu_3Pd and Cu_3Pt alloys.

Another type of ordered structure, that of Fe_3Al, is shown in Fig. 2 with a graphical representation of the distribution of atoms among the phase sites determined by Bradley and Jay.[6] At compositions up to 18 atomic percent Al the distribution is uniform among the four sites (in the larger unit) for both quenched and annealed alloys. When more numerous, the Al atoms in the annealed alloys tend to segregate into the *b* positions; at 25 atomic percent Al, 92 percent of the Al atoms occupy *b* positions, the remaining 8 percent being distributed among the *a*, *c* and *d* positions. At 25 atomic percent Al, small cube centers are so occupied by Al atoms that on traversing the lattice along cube diagonals one meets Al atoms in alternate cube centers. As Bragg[7] has pointed out, the Al atoms tend to keep as far apart as possible. On passing to higher Al compositions more Al atoms go into the remaining *b* positions and also into *d* positions. At about 40 atomic percent Al the aluminum atoms are equally distributed among the *b* and *d* positions. At 50 atomic percent one obtains a CsCl structure with Al atoms occupying the cube centers and Fe atoms the corners. Phragmén[8] has observed a similar superstructure in Fe_3Si. More complicated superstructures are found in Mg_3Cd,[9] Ni_3Sn[10] and Cr_2Al.[11]

In general, a disordered alloy persists in its random state on cooling from elevated temperatures down to a critical temperature T_c, at which point the atoms begin to distribute themselves in an orderly manner among the phase sites. The degree of order (long distance order, see below) increases with decreasing temperature down to a point where diffusion can no longer take place. At temperatures below this point the alloy can be pictured as "frozen" into a definite state.

Elements of Superstructure Theory

Long distance order can be defined as the orderly arrangement of atoms among atom sites. This can easily be illustrated in the case of an ordered structure of β-brass. In the disordered alloy the body-centered cubic lattice may be divided into two simple cubic lattices, the points of one lying at cube centers in the other. In the

ordered alloy these two sites are occupied by copper and zinc, respectively. So also, in the Cu_3Au ordered structure already described, the gold atoms occupy one simple cubic lattice and the copper atoms the remaining three simple cubic lattices to form the face-centered cubic structure (Fig. 1B).

In order to express the degree of long distance order quantitatively we shall, following Bragg and Williams,[12] let n denote the number of atom positions which may be occupied by either of the two kinds of atoms (A and B) forming the alloy. Ordinarily, n is equal to the total number of atoms in the alloy. Of these n disposable sites a fraction r would be occupied by A atoms in the state of perfect order. Call these α-sites. Call the rest, $(1-r)n$ in all, β-sites. In the state of perfect order the probability p that an α-site is occupied by an A atom is unity, whereas at temperatures great enough to permit atomic interchange of A and B atoms p is less than unity. When the lattice is completely disordered p will simply be equal to r. The degree of order is defined as

$$S=\frac{p-r}{1-r}.$$

When order is complete, p is unity, and S is therefore 1. For complete disorder $p=r$ and $S=0$.

Attempts to explain the presence of the critical temperatures, sometimes called Curie temperatures of order, where the alloys begin to take on an ordered structure on cooling or where the ordered structure passes into the disordered state on heating, have been made on general thermodynamical, and classical statistical bases. The first statistical treatment appears to have been given by Gorsky,[13] Borelius,[14] Dehlinger[15] and others have used a more general thermodynamical method. In the following brief summary of the subject we shall discuss the assumptions and results in later work by Bragg and Williams and shall add a few additional remarks on the general theory as modified by considerations of local order. Local or short distance order was first considered by Bethe for alloys of equal atomic compositions and his argument was later extended by Peierls along similar lines to cover the composition ratio 3 : 1.

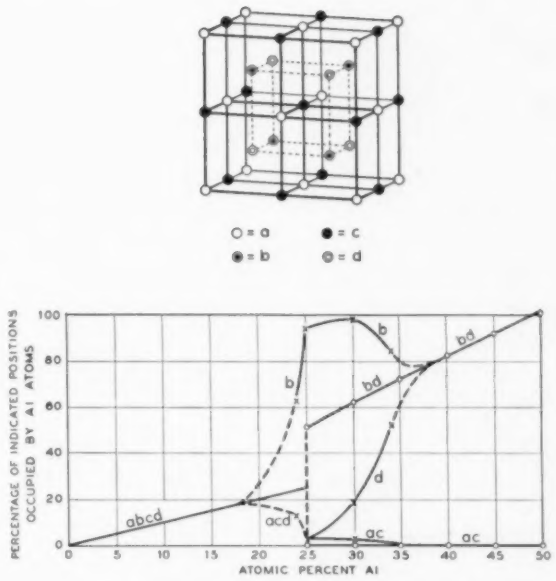

FIG. 2. Distribution of aluminum atoms among the four indicated positions a, b, c, d in the Fe-Al system. Full line up to 18 percent Al represents distribution for both rapidly cooled and annealed alloys. From 18 to 38 percent Al lines marked (0) refer to cooled alloys, and lines marked (X) refer to annealed specimens. The line from 38 to 50 percent Al refers to both rapidly cooled and annealed alloys.

Bragg and Williams[12, 16, 17] considered the distribution of the A atoms among the α- and β-sites to be governed by the Boltzmann factor, and postulated an ordering energy V, which is the energy necessary to interchange an A atom in an α-site with an adjacent B atom in a β-site. The ordering energy V at any given temperature T determines the distribution of the A and B atoms among the α- and β-sites. Most generally, therefore: $S=S(V, T)$. With a given ordering energy V the degree of order S can be obtained as a function of the temperature. A plot of S *versus* T with $V=$const. displays a smooth exponential decrease of S with increasing temperature with no indications of a Curie point of order. To make the theory consistent with experiments we make the additional plausible assumption that the ordering energy is a function of the degree of order and that $V=V_0S$, where V_0 is a constant. We shall further assume that V is independent of the temperature at least to a first approximation. Thus V_0 is the ordering energy at $S=1$ or for perfect order. We shall call the equation $V=V_0S$ our $V(S)$ relationship.

We now have two relationships between S and

V, $S(V, T)$ and $V(S)$. Their simultaneous solution for a series of values of T gives S as a function of T. Curve A in Fig. 3 is the theoretical curve for the case of equal atomic proportions, e.g., β-brass. In the case of the atomic ratio 3 : 1 e.g., Cu_3Au, the degree of order suffers a precipitous jump at T_c (curve B). The experimental results are in fair agreement with the theoretical predictions, but the alloys tend to disorder more rapidly on heating near T_c than the theory predicts.

For the case of equal atomic proportions it is easy to obtain T_c in terms of the ordering energy V_0 at $S=1$, *viz.* $V_0=4KT_c$. This relationship can alternatively be used to obtain rather crude values of V_0 from experimentally determined values of T_c.

The present theory completely fails to account for the variation of T_c for composition ranges near $r=\frac{1}{4}$ (e.g., Cu_3Au) general shape of the T_c *vs.* composition curve in the vicinity of $r=\frac{1}{2}$ (e.g., Fe_3Al) is in reasonable agreement with theory.[16]

A test of the above theory can be obtained by comparison of theoretical and experimentally determined energy changes accompanying the order-disorder transformations. One can easily show that the total energy change in the specimen in changing from complete disorder to a degree of order S is

$$E_0-E_s=\frac{V_0nr(1-r)S^2}{2},$$

which reduces to $E_0-E_s=(V_0n/8)S^2$ when $r=\frac{1}{2}$. Substituting the value $V_0=4KT_c$ also valid when $r=\frac{1}{2}$ we obtain

$$E_0-E_s=\frac{nKT_cS^2}{2}=\frac{RT_c}{2}S^2,$$

which gives for a change from disorder to complete order, from $S=0$ to $S=1$,

$$\frac{E_0-E_1}{RT_c}=0.5.$$

Figure 4 gives the curve of specific heat *vs.* temperature as determined experimentally by Sykes and Jones[18] for the Cu_3Au transformation. The theory predicts the general shape of the curve, but does not explain anomalously large specific heat above T_c. Figure 5 depicts the energy of transformation, for Cu_3Au, in calories per gram as a function of temperature derived from Fig. 4, and for comparison the theoretical curves of Bragg and Williams and of Peierls. The experimental values fall between the two theoretical curves. These experimental results also confirm one rather striking prediction of the theory, the existence of a latent heat for an alloy with 3 : 1 atomic ratio. Fowler[19] has pointed out that this instance represents the first known example of a latent heat occurring in a single homogeneous phase.

It is assumed in the theory of Bragg and Williams that the ordering energy is proportional to the long distance order S prevailing throughout the crystal and is independent of local variations in S which undoubtedly exist. To take into consideration these local variations Bethe[20] introduced the conception that the ordering energy for a given atom is a function of the degree of order of its nearest neighbors. To evaluate the configurational energy depending on the order of nearest neighbors, Bethe introduces a parameter $\frac{1}{2}(1+\sigma)$ somewhat analogous to the "p" of Bragg and Williams theory. In the latter theory we were interested in whether a certain α-site was occupied by an A or by a B atom. In Bethe's theory we are interested in knowing how a given atom is surrounded: i.e., $\frac{1}{2}(1+\sigma)$ is the probability that any given atom

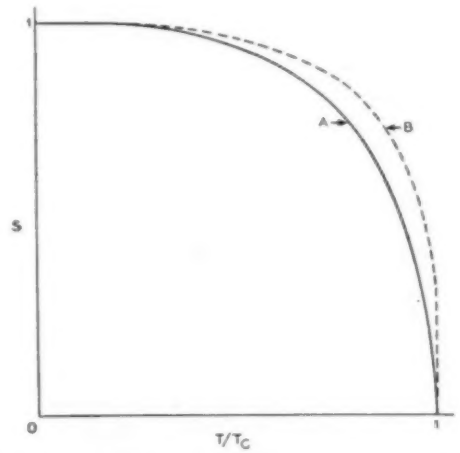

FIG. 3. Equilibrium degree of order *vs.* temperature. A for $r=\frac{1}{2}$, B for $r=\frac{1}{4}$.

will have the right kind of neighbors. If σ is known we can then calculate for any given alloy the number of like and unlike pairs of atoms, and hence the configurational energy. In order to calculate σ we consider a small piece of the crystal consisting of one α-site and Z nearest neighbors. We then replace the rest of the crystal by an ordering force dependent on the degree of long distance order in that outer portion. Bethe's theory predicts that at a certain critical temperature T_c long distance order will vanish, and with it the external ordering force. However, σ does not vanish, but merely decreases with increasing temperature, i.e., at temperatures above T_c there still exists a certain probability that a given atom will be surrounded by "right" neighbors. It follows then

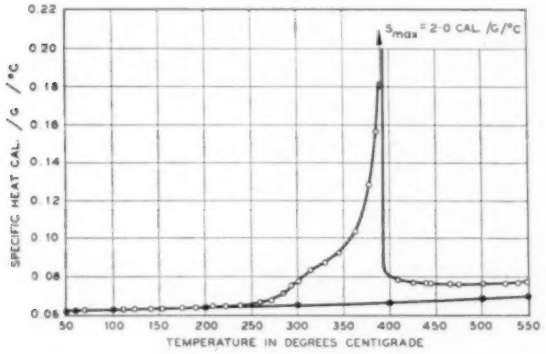

FIG. 4. The variation of specific heat with temperature of a Cu_3Au alloy is shown by the curve marked (0). The curve marked (●) is calculated from the specific heats of Cu and Au assuming a pure mixture.

from Bethe's theory that there should exist some configurational specific heat at temperatures above T_c. This prediction is in good agreement with the experimental results of Fig. 4. Peierls[21] extended the calculation of Bethe to the case of 3 : 1 atomic proportions. His theoretically predicted curve for the energy of transformation of Cu_3Au is given in Fig. 5.

Methods for Measuring the Degree of Order

The degree of order can be ascertained at least in principle by measuring electrical conductivity, magnetic susceptibility, intensity of the superstructure lines, or crystallographic constants. The effects of order on electric and magnetic properties will be discussed. An example of crystallographic effects is furnished by

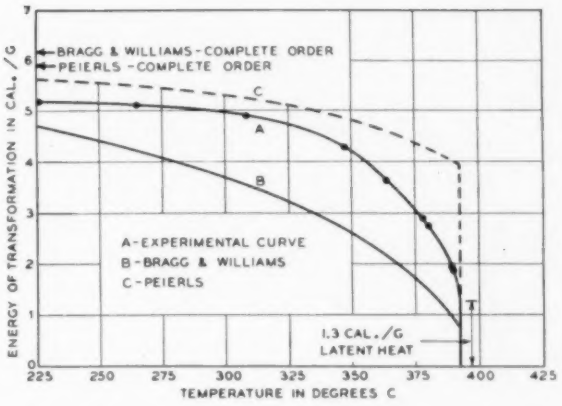
BRAGG & WILLIAMS - COMPLETE ORDER
PEIERLS - COMPLETE ORDER
A - EXPERIMENTAL CURVE
B - BRAGG & WILLIAMS
C - PEIERLS
1.3 CAL./G LATENT HEAT
ENERGY OF TRANSFORMATION IN CAL./G
TEMPERATURE IN DEGREES C

FIG. 5. Energy of transformation *vs.* temperature.

Gorsky's[13] measurements of the axial ratio c/a for the tetragonal phase of CuAu. He assumes the "tetragonality" to be provoked by the ordering of the structure. For this ordered structure the axial ratio c/a is 1.065 at the initial stage of the ordering process and increases with increasing degree of order to 1.085 for the greatest degree of order obtainable.

Effect of Order on Electrical Resistivity

The transformation of an alloy from the disordered condition to one possessing long distance order is accompanied in many cases by pronounced changes in electrical conductivity. Fig. 6 shows a typical curve of electrical resistivity *vs.* temperature obtained from measurements[22] on a Cu_3Au alloy. The magnitude of the initial decrease at T_c (381°C for Cu_3Au) and the general shape of the curve functions depend upon the cooling rate. A curve taken at a rate sufficiently slow to permit the attainment of equilibrium at all temperatures would be the limiting curve. The solid line in Fig. 6 represents equilibrium conditions for temperatures above 350°C. For faster rates of cooling, too fast to allow the alloy to relax into its equilibrium state, one obtains similar curves which fall between this curve and the dashed line obtained after quenching. A plot of resistivity *vs.* temperature for this alloy on reheating at the same rate at which it was cooled, invariably falls below the curve obtained on cooling. An alloy cooled rapidly by quenching from a temperature well above 381°C (T_c) in many cases will approximate the same order as that prevailing at the

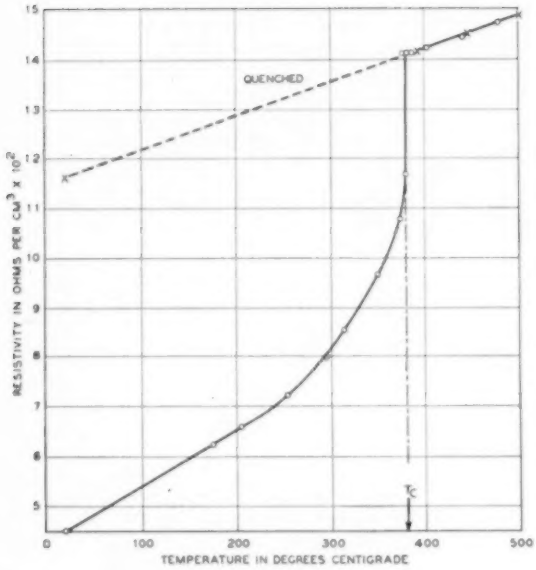

FIG. 6. Electrical resistivity *vs.* temperature for Cu_3Au. Cooling rate sufficiently slow to permit attainment of equilibrium at temperature above 350°C.

temperature of quench. The resistivity of such an alloy is found to possess a value obtained by extrapolating to room temperature the curve of resistivity *vs.* temperature for temperatures above T_c.

Figure 7 shows the results obtained from resistivity measurements[4] made on a series of copper-gold alloys. They include (*a*) disordered alloys obtained by rapidly cooling from 650°C, (*b*) alloys subjected to prolonged annealing at 200°C where an ordered state prevails in certain composition domains, (*c*) alloys obtained by slow cooling in an annealing furnace, which provokes partial ordering in composition regions known to form ordered structures. The plot of resistivity *vs.* composition for the disordered alloys (*a*) yields a smooth curve with a maximum at 50 atomic percent Au. A similar plot of resistivity *vs.* composition for the series (*b*) annealed at 200°C displays radical changes of resistivity in composition ranges where ordering has been directly confirmed by x-rays. The curve possesses minima at 25 and 50 atomic percent Au. An alloy of the latter composition suffers a decrease of resistivity, on annealing, from 14.1 to 3.63 micro-ohm-cm. This general behavior in resistivity is typical of systems containing regions which order on annealing, and may be used with quite a degree of assurance as a criterion for the presence of an ordered region. Fig. 8 shows similar measurements[23, 28] on the system copper-palladium. The change in resistivity on ordering for an alloy containing 48 atomic percent Pd was found to be from 36 to 3.6 micro-ohm-cm, the latter being below that for pure palladium. There is a striking difference between the results obtained for Cu-Au and Cu-Pd systems. In the former the plot of resistivity *vs.* composition for the annealed alloys displays a striking degree of symmetry around the Cu_3Au and CuAu compositions whereas for alloys subjected to similar treatment in the Cu-Pd system we note that the plot is decidedly asymmetrical around both Cu_3Pd and CuPd. An alloy containing 49 or 50 atomic percent Pd cannot be transformed into an ordered structure even on annealing for periods of several weeks. The presence of an appreciable degree of short distance order does not appear to effect the electrical resistivity.[18]

Effects of Order on Mechanical Properties

The ordering process is accompanied by distinct changes in mechanical properties. Young's modulus[24] increases on ordering in the Cu_3Au,

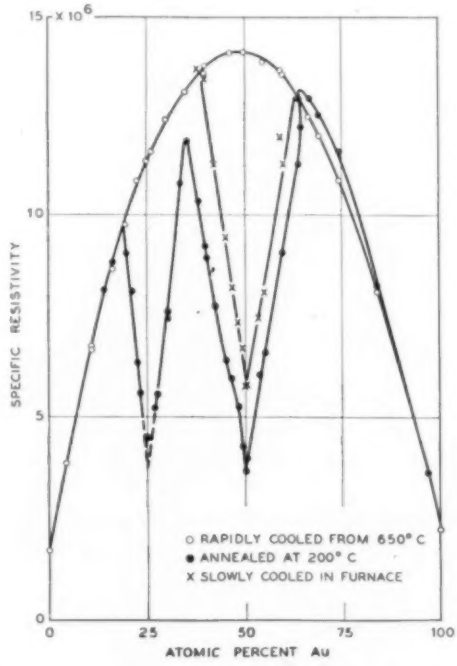

FIG. 7. Electrical resistivity *vs.* composition for the Cu-Au system.

Cu_3Pd and Cu_3Pt phases, whereas it suffers a decrease on ordering in CuAu and CuPd alloys. Mechanical tests[25] such as hardness, tensile strength and elastic limit all show an increase on ordering. A Cu-Au alloy[26] containing 50 atomic percent Au, quenched from an elevated temperature to retain the disordered state, was found to increase in hardness about 90 percent on annealing at temperatures from 150 to 300°C. A plot of hardness *vs.* time for this alloy so treated displays characteristics strikingly similar to those obtained in precipitation-hardening alloys. For instance, an anneal at 150°C shows the alloy continuing to increase in hardness after 20 minutes of anneal, whereas a few minutes anneal at a temperature of 300°C provokes a maximum in hardness which decreases with

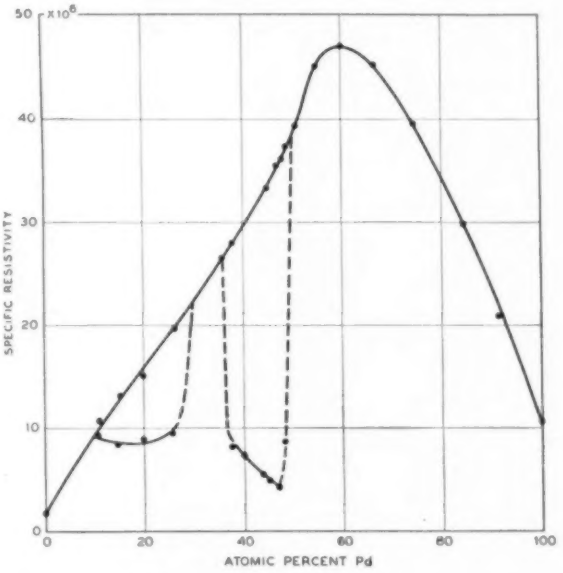

FIG. 8. Electrical resistivity *vs.* composition for the Cu-Pd system.

further anneal. The hardness in the final state, highly ordered, is greater than that of the disordered alloy. The Cu_3Au alloy[25] has also been found to possess this "age-hardening" characteristic to a slight extent.

Effects of Order on Magnetic Properties

Dia- and Paramagnetic Alloys

The full line in Fig. 9 shows how the diamagnetic susceptibility depends on the composition for disordered substitutional isomorphous series

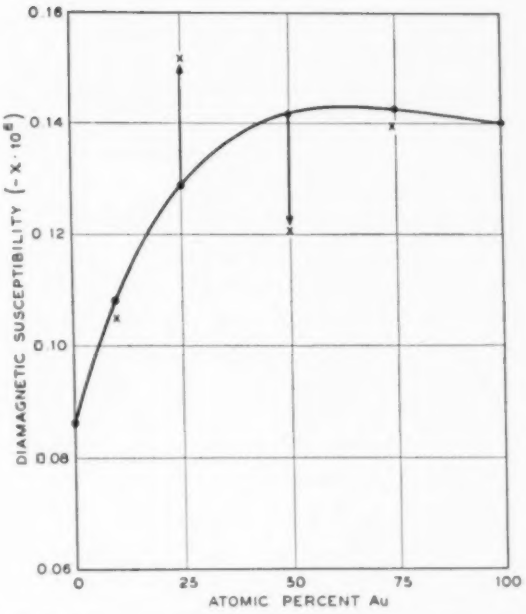

FIG. 9. Diamagnetic susceptibility *vs.* composition for the Cu-Au system. Arrows indicate susceptibility for ordered alloys.

of solid solutions of the two weakly diamagnetic metals copper and gold. It was first shown by Seemann and Vogt[27] that the formation of an ordered structure at compositions of 25 and 50 atomic percent gold is accompanied by a pronounced change in the diamagnetic susceptibility amounting in some cases to as much as 19 percent. However, the direction of the change was different in two cases; the Cu^3Au alloy suffered an increase, whereas the CuAu alloy displayed a decrease of the same order of magnitude in the diamagnetic susceptibility. Fig. 10 depicts a plot of magnetic susceptibility *vs.* composition for the copper-palladium isomorphous series of solid solutions.[28] In this series we have an example of the effect of alloying the strongly paramagnetic metal palladium with the weakly diamagnetic one copper. Small additions of copper suffice to effect a pronounced lowering of the paramagnetic susceptibility from that of pure palladium the alloys containing more than 50 atomic percent copper being diamagnetic. It is surprising to find that small additions of the strongly paramagnetic palladium to the weakly diamagnetic copper provoked an increase in diamagnetic susceptibility up to a concentration of about 30 atomic percent Pd, at which point the curve decreased and crossed over into the paramagnetic state at

ca. 50 atomic percent Pd. Similar behavior is also found in the systems Ag-Pd and Au-Pd.

Alloys in both the Cu_3Pd and CuPd regions suffer an increase in diamagnetic susceptibility on ordering. It is to be noted that on ordering the diamagnetic susceptibility of Cu_3Pd and Cu_3Au are both increased, whereas CuPd and CuAu are changed in opposite directions. It is perhaps of some interest to point out that the ordered Cu_3Pd alloys possess a minimum in magnetic susceptibility at about 17 atomic percent Pd, the composition possessing the maximum degree of order as determined from intensity of superstructure lines as well as by conductivity measurements. In the CuPd region, the greatest change in the magnetic susceptibility on ordering is observed at about 38 atomic percent palladium, whereas the alloy possessing the lowest resistivity contains 47 atomic percent palladium. The ordering process is in every case accompanied by a pronounced lowering in resistivity.

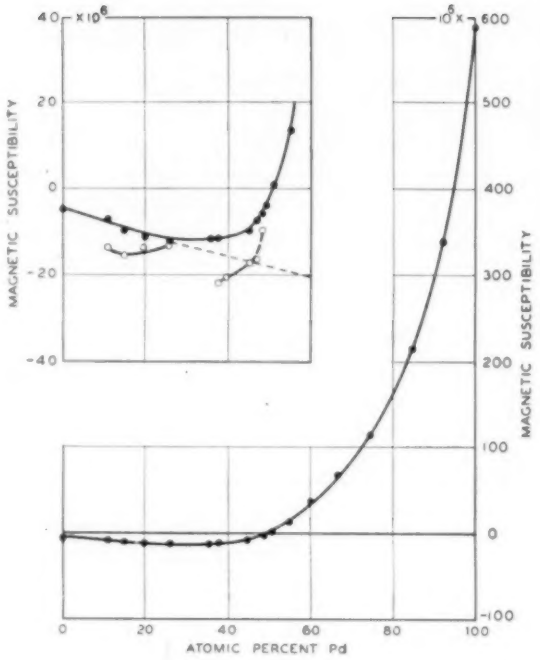

FIG. 10. Magnetic susceptibility *vs.* composition for the Cu-Pd system. The empty circles represent susceptibility of ordered alloys.

Ferromagnetic Alloys Containing No Ferrous Component

It has been shown by Bradley and Rodgers[29] as well as by Heusler[30] and others that the presence of ferromagnetic properties in the Cu_2AlMn alloy is associated with a state of order prevailing in this alloy. The copper in it can be replaced by silver, or the aluminum by tin, without destroying the ferromagnetism.

Among other nonferrous alloys displaying ferromagnetic properties which appear to bear some relationship to a degree of order is the interesting example of the platinum-chromium system.[32] In the region from 20 to 50 atomic percent chromium the alloys become ferromagnetic and the curve of saturation magnetizations *vs.* composition has a maximum at *ca.* 30 atomic percent Cr. Moreover, the saturation magnetization is a function of heat treatment; the annealed alloys possess considerably higher values than the rapidly cooled ones. The crystal structure of alloys in the ferromagnetic region is that of platinum and there is no direct evidence of the presence of a superlattice until a chromium content of some 40 atomic percent is attained. However, the lines of the x-ray diffraction pattern become somewhat fuzzy before this Cr composition is reached. The intensity of the superstructure lines increases with increasing chromium content even above 50 atomic percent and is much greater in annealed than in rapidly cooled alloys. It is thought, although the evidence is not conclusive, that these superstructure lines come from the Cr_3Pt region. Thus it may be that the ferromagnetic characteristic occurs in a region of the Pt-Cr system where only partial order exists, and disappears at some critical degree of order.

Many other nonferrous alloys not containing a ferromagnetic component display ferromagnetism, but no evidence exists which correlates their behavior with a characteristic ordered structure. Such alloys[31] are $SnMn_2$, $SnMn_4$, $MnSb_2$ and Mn_2Sb.

Ferromagnetic Alloys Containing at Least One Ferrous Component

The system Fe-Pd is an example of a binary alloy containing a ferromagnetic component, in which there is evidence that development of superstructure changes the magnetic properties. Jellinghaus[33] reports that in the FePd region a quenched alloy containing 50 atomic percent Pd

possessed a coercive force of two oersteds. On annealing for 15 hours at 500°C the coercive force increased to 260 and on a further anneal of 15 hours it dropped to 190. On still further annealing at 500°C for some 88 hours the value of the coercive force reached a final and steady value of 150. X-ray diffraction patterns of this alloy showed the quenched specimen to possess a random face-centered cubic structure like that of pure Pd. The alloy after the first anneal of 15 hours displayed no appreciable change in the x-ray pattern other than a slight broadening of

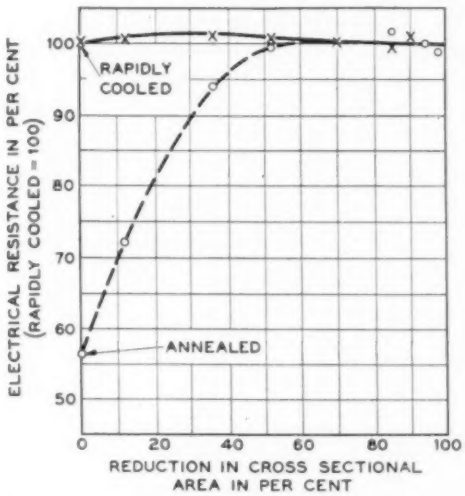

FIG. 11. Effect of cold work on a Cu_3Au alloy. "Annealed" and "rapidly cooled" refer to thermal treatment preceding cold work.

the (311) lines, whereas the additional anneal of 15 hours which decreased the coercive force to 190 oersteds provoked a change from the random face-centered cubic to an ordered tetragonal structure similar to that of the ordered CuAu. The axial ratio in this case is $c/a = 1.03$, somewhat smaller than the ratio observed in the similar CuAu ordered phase. Unfortunately this evidence of Jellinghaus is not in agreement with that of other observers[34] who report no evidence of the FePd superstructure, though they find an ordered phase in Fe_3Pd.

In the similar Fe-Pt system interesting magnetic properties are observed in the region from 26 to 70 atomic percent Pt. Graff and Kussmann did not observe an ordered phase in the system; the face-centered cubic phase stable at elevated temperatures transforms into a body-centered phase stable at low temperatures. These authors report a coercive force of 1750 oersteds for quenched alloys, which on annealing at 500°C decreases to about 500 oersteds. Jellinghaus[33] on the other hand reports that an FePt alloy in which five atomic percent of Pt is replaced by rhodium, is an ordered CuAu type structure possessing a coercive force of 1400 oersteds and a remanence of 3700 gauss. It is possible that the presence of the rhodium is responsible for the appearance of the face-centered tetragonal ordered phase of the CuAu type.

It is also of interest to mention that a commercial permanent magnetic material called Mishima alloy, containing Fe, Ni and Al, when subjected to a proper heat treatment attains a state of extreme magnetic hardness which is accompanied by a superstructure.[35] The degree of order, however, as measured by the intensity of the superstructure lines, is not at all proportional to the magnetic hardness.

Fallot[36] has reported interesting magnetic measurements on the alloy series Fe-Al, Fe-Si, etc. which he considers to be evidence for the existence of superstructures at compositions other than the well-known Fe_3Al and Fe_3Si,

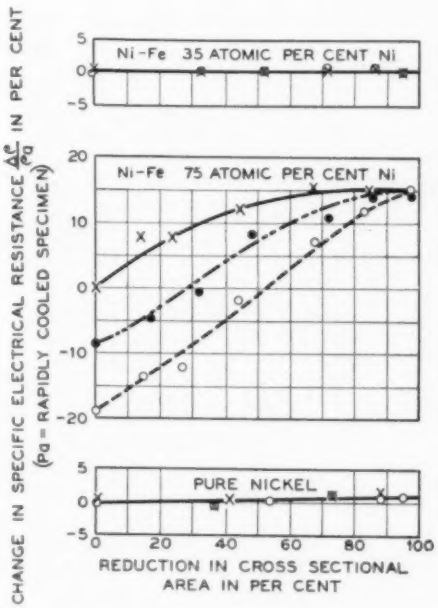

FIG. 12. Effect of cold work on pure Ni, and Fe-Ni alloys containing 35 and 75 atomic percent Ni. "Rapidly cooled" etc. refer to thermal treatment preceding cold work. Crosses, rapidly cooled and cold worked; circles, annealed and cold worked; black dots, furnace cooled.

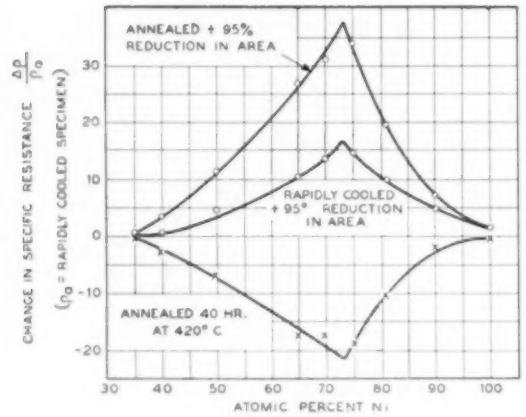
ANNEALED + 95% REDUCTION IN AREA
RAPIDLY COOLED + 95° REDUCTION IN AREA
ANNEALED 40 HR. AT 420° C
CHANGE IN SPECIFIC RESISTANCE $\frac{\Delta\rho}{\rho_0}$ (ρ_0 = RAPIDLY COOLED SPECIMEN)
ATOMIC PERCENT Ni

FIG. 13. The upper curves depict variation with composition of the change in resistance, provoked by cold work for alloys which had been annealed and rapidly cooled respectively, before cold work. The lower curve depicts the difference in resistance between annealed and rapidly cooled specimens.

namely at Fe_7Al at Fe_7Si. Such measurements must be confirmed by other more direct means before the presence of superstructures at these compositions can be acknowledged. But Fallot's method may prove to be of value in studying ordered phases in alloy systems where the constituent atoms possess similar scattering power for x-rays.

Effect of Cold Work on Order

The effect of cold work on order has been most extensively investigated by Dahl[37] for the Cu-Au system. Fig. 11 depicts the effect of cold work on the resistivity of a Cu_3Au alloy in the ordered and the disordered state. The resistance of the disordered alloy increased some, only about 2 percent, after the most severe cold working. This behavior is typical for disordered solid solutions and pure metals. In the figure the lower resistivity curve shows the effect of the same degree of cold work on a Cu_3Au alloy initially in the ordered state. The resistance increased with increasing degree of cold work and at 60 percent reduction in cross-sectional area attained the same resistance as that of the random solid solution. X-ray diffraction patterns showed that the increasing degree of cold work was accompanied by a destruction of the ordered arrangement as evidenced by a weakening of the superlattice lines and their final disappearance at 60 percent reduction in area. Dahl then carried out a similar series of experiments on alloys of the Fe-Ni system. Fig. 12 shows the effect of cold work on alloys containing 35 and 75 atomic percent nickel, and on pure nickel. For the Ni_3Fe alloy the increase in resistance with increasing degree of cold work was found to be greater than that to be expected were the alloy a random solid solution. The largest effect was observed on alloys originally annealed for thirty-six hours at 420°C. The pronounced similarity between the Ni_3Fe and the Cu_3Au alloys indicates, although it does not conclusively prove, that the former may also be considered as an ordered lattice. The effect on the quenched alloys can be explained as due to a partial ordering which occurred during the rapid cooling. The change in resistivity on cold working in the Fei-Ni system is given as a function of composition for annealed and rapidly cooled alloys in the upper two curves in Fig. 13. The largest effect is observed near 75 atomic percent Ni. From the same figure it is seen that the difference in resistivity of a series of annealed

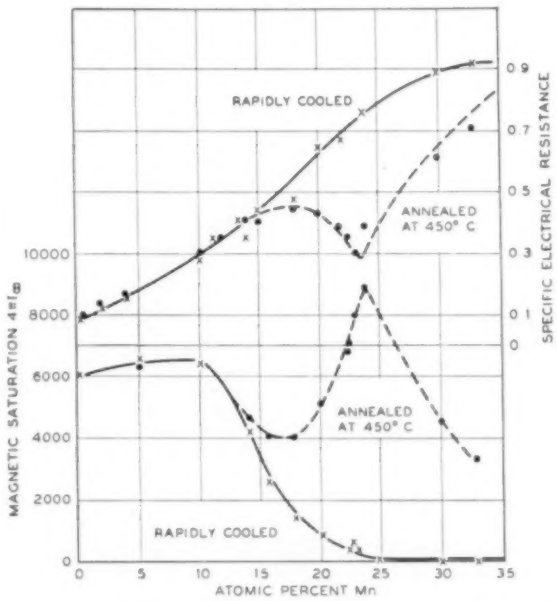

FIG. 14. Variation with composition in the magnetic saturation ($4\pi I_{\infty}$) and electrical resistance of Ni-Mn alloys.

alloys, relative to the same alloys in the rapidly cooled state, is a maximum at about the same composition. It is perhaps of some significance to note that several magnetic changes occur in this composition range, e.g., the maximum in the initial permeability curve as a function of com-

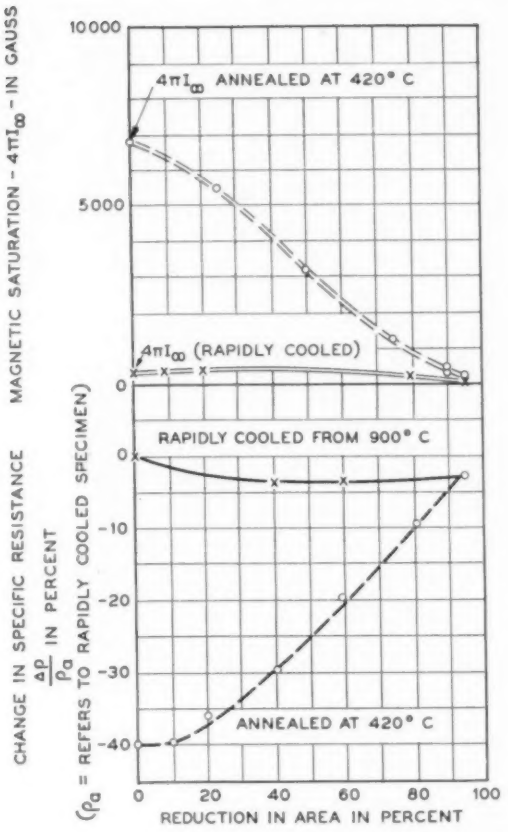

FIG. 15. Effect of cold work on the magnetic saturation ($4\pi I_\infty$) and electrical resistance for a Ni_3Mn alloy.

position, change in sign of the magnetostriction, and a maximum in the curve of Curie point *vs.* composition. In this region also the magnetic anisotropy vanishes.

Figure 14 depicts the relation as determined by Kaya and Kussmann,[38] between electrical resistance, magnetic saturation intensity ($4\pi I_\infty$) and composition for a series of rapidly cooled and of annealed Ni-Mn alloys. The minimum in the resistance *vs.* composition curve at *ca.* 25 atomic percent Mn is indicative of an ordered arrangement in the annealed alloys. Also magnetic saturation curve displays a maximum at this composition for the alloys in the same annealed condition. As for Ni_3Fe, Dahl[37] again studied the effect of cold working on the resistance and the magnetic saturation intensity for the Ni_3Mn alloy. The results are presented in Fig. 15. Ni_3Mn, although not ferromagnetic in the rapidly cooled condition, possesses in the annealed state a $4\pi I_\infty$ value of *ca.* 8500 gauss, somewhat higher than that of pure nickel. With cold working, $4\pi I_\infty$ suffered a pronounced decrease, and the alloy became nonferromagnetic after extreme cold working. The resistivity of annealed and quenched alloys suffered a change with cold working in much the same manner as the Cu_3Au alloys. In order to show that the behavior described for the Ni_3Fe and Ni_3Mn alloys cannot be due to a precipitation provoked by cold working, a Ni-Fe alloy containing 40 atomic percent Ni and 1.5 percent Be, known to be an age-hardenable alloy of the precipitation type, was subjected to the same treatment as we have described for the Ni_3Fe and Mn_3Ni alloys. It was found that the resistivity of the rapidly cooled and annealed alloys changed similarly with hard working, in contrast to the behavior of the Ni_3Mn alloy for which the rapidly cooled and annealed alloys behaved very differently as shown in Fig. 15.

It is generally true that an alloy has different lattice constants in the ordered and disorder states. Measurements[39] on lattice constants for annealed and rapidly cooled Fe-Ni alloys near 75 atomic percent Ni failed to show any difference due to heat treatment. Thus the evidence for superstructure in the Fe-Ni alloys is conflicting.

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The history of scientific discovery and the application of scientific knowledge to human activities in every field reads like a romance, and I can imagine no more interesting career for anyone whose tastes lie in that direction. The interest and importance of the scientific career, however, are not confined to the laboratory or the classroom, but should be regarded as a major, if indeed not the major, creative influence on this generation. In national economic well-being, especially in making high standards of living possible, in the evolution of both the powers and forms of national institutions, in the efforts to create and secure social harmony and co-operation, the scientific method, if followed, would be of great assistance and would save some futile experiments, mistaken agitation and unworkable proposals.

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High Rotational Speeds

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IT is a general historical fact that, following each improvement in experimental technique, rapid progress has been made in the fields of science to which it was applicable. Therefore it may be worth while to review a few of the recent advances made in the production of high rotational speeds because of their possible use in many different types of research. With the exception of Svedberg's, most of the recent successes in the production and use of ultra-high rotational speeds have resulted from the employment of one or a combination of the three following devices: the air or gas bearing, the flexible shaft and the oil or liquid sealed vacuum gland. All of the principles upon which the above are based have been known for a long time but they have been successfully applied to the problem under review only in the last few years.

For many generations of students almost every lecturer in elementary physics has shown his class a ball freely supported upon a jet of air. The ball could be made to remain almost stationary, to move, or to spin, depending upon the wishes of the experimenter. Among other things this demonstration is usually made to illustrate the well-known principle of Bernoulli. However, it was not until 1925 that Henriot and Huguenard,[1] by means of a very ingenious arrangement, successfully utilized the general principles involved in the above experiment to spin small cone shaped rotors to very high speeds in air at atmospheric pressure. Since this original work of Henriot and Huguenard their method has been modified and improved by a number of workers[2–8] until at the present time the rotors are extremely stable. For example, a rotor may be loaded or unloaded while at full speed or made to carry a superstructure which need not be dynamically balanced with extreme care. In this review, space does not permit a full description of the original method of Henriot and Huguenard followed by the modifications which have been introduced in their chronological order. (For this see original references in Bibliography.) Instead, a brief description will be given of a general purpose apparatus that has been found very satisfactory.[7, 8]

Figure 1b is a cross section drawing and Fig. 2 a picture of the apparatus. Compressed air, admitted to the stator cup through flexible tubing, flows through the straight holes or channels LL' (Fig. 1a) and impinges upon the flutings of the rotor. As a result the rotor is both lifted off the stator and given a spin. The Bernoulli forces developed by the motion of the air between the rotor and stator prevent the former from flying out of the latter and, together with the weight of the rotor, balance the forces of the air impinging upon the rotor. This automatic balance takes

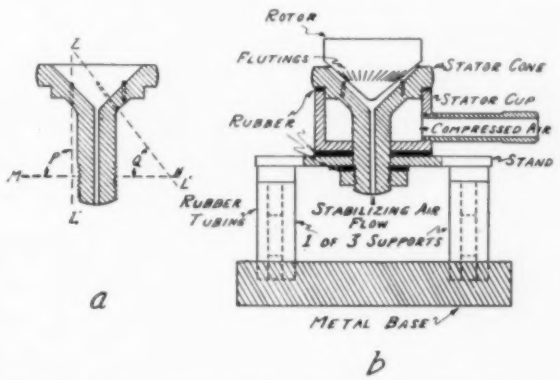

FIG. 1. *a*, Cross-sectional drawing of stator cone; *b*, cross-sectional drawing of stator and support with rotor in place.

place when the rotor is only a fraction of a millimeter above the stator so that the rotor rides upon a very thin cushion of air. In order to increase the stability of this air cushion or "bearing," additional air is allowed to flow from the atmosphere into the vertex of the stator cone (labeled stabilizing air flow, Fig. 1b). In some work[3] this stabilizing air has been forced into the stator under the desired pressure above atmospheric, or a valve[3, 6] has been added to limit the amount of air flow. However, with the dimensions given in Fig. 1b these added complications are unnecessary as the rotor is extremely stable at all usable speeds with the channel simply open to the atmosphere as shown. For example, a rotor can be loaded by suddenly pouring liquid into it while it is spinning, or the driving air pressure can be changed suddenly from 5 to 150 lb./in.^{2} or *vice versa* without disturbing the rotor's stability. In Fig. 1, $P=90°$; $Q=45°$ to $46°$; angle of stator cone $=91°$ to $92°$; angle of rotor cone $=102°$ to $103°$. The upper openings of the 5 to 9 directed channels LL' (bored with no. 65 twist drill in Fig. 2) lie in a circular section of the stator cone approximately midway between vertex and base. The diameter of the base of the stator is $1\frac{1}{8}$ inches. However, the number and size of the channels LL' may be varied over wide ranges. From 3 to 20 channels bored with twist drills of sizes from 45 to 76, depending upon the size of the rotor and stator, have been used successfully. The stator can be built to fit any size of rotor by simply changing all dimensions proportionately. An investigation of the efficiency of different types and directions of channels and rotor flutings has been carried out at the University of Virginia by Garman.[5] The stator should be flexibly mounted because, at certain rotational speeds, the air jets impinging upon the rotor, asymmetries in the rotor, etc., set up resonance vibrations which may be communicated to the stator. Usually the rotor can be accelerated through these resonance frequencies so quickly that they cause no serious trouble, but it is always preferable to keep them damped in the stator mounting, say by rubber or cork. Also it is important to avoid vibrations in the air supply line or stator box caused, for example, by a vibrating valve, constriction in nonflexible tubing, or wrong type of jet opening. A novel way of mounting the stator on flexible metal tubes which take the place of LL' and avoid the stator box has been used by Garman[5] and Girard and Chukri.[6] Fig. 3 shows another satisfactory stator mounting. For the various types of mountings and methods of construction of the various parts of the apparatus reference should be made to the original papers.[1–8] The stators can be made of any easily machinable materials such as steel, brass or Duralumin. However, care must be taken in selecting the rotor material because the bursting strength of the rotor often determines the maximum attainable rotational speed. This subject will be referred to again later in this review. The property of the air bearing of allowing the rotor to seek its own axis of rotation is exceedingly

FIG. 2. Photograph of rotor, stator, and stator cone.

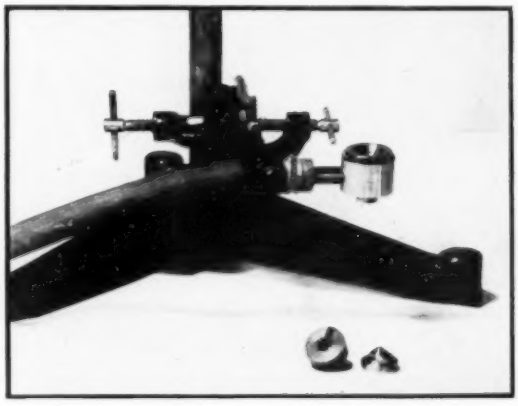

FIG. 3. A simple stator mounting. The stator and rotor (with flutings up) were used to make curve E of Fig. 5.

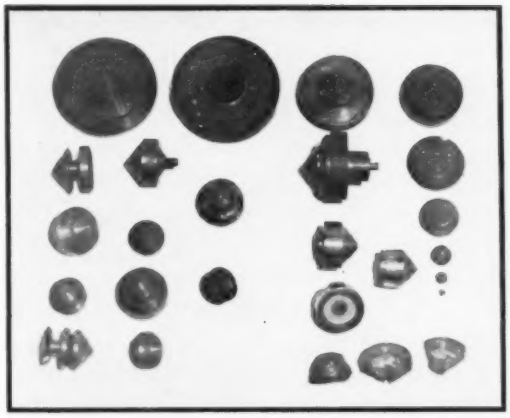

FIG. 4. Miscellaneous collection of rotors. At bottom right are some pieces of exploded rotors.

useful as this not only makes it possible to spin a wide variety of shapes and sizes of rotors (Fig. 4) but also simplifies the mounting of apparatus upon the rotor.

The above method is suitable for spinning almost any size or shape of rotor in air or other gases at about atmospheric pressure. However, as is well known, the air friction on a surface increases very much with its speed so that the power input must be greatly increased with size and speed of rotor. This is well illustrated by the curves of Fig. 5. By using hydrogen instead of air both to drive and surround the rotor the friction is not only reduced but the gas in the jets strikes the rotor with greater speed for a given pressure. For example, with a 9 mm diameter rotor driven and surrounded by hydrogen, we have obtained 21,600 r.p.s. and hence centripetal acceleration in excess of 8 million gravity. With smaller rotor and stator this could probably be much increased.

The above type of apparatus has been used quite successfully for spinning small mirrors to very high speeds.[1, 3, 9–11] Fig. 6 shows a spark photograph taken at Virginia with such a rotating mirror. Two events occurring within a time of the order of 10^{-9} sec. can be resolved. Also the apparatus has been used by H. W. Beams, R. L. King and others [12–16] as a centrifuge in many biological experiments where precise temperature control or thermal equilibrium throughout the centrifuge is not essential. Harvey[16] and Pickels[17] have devised simple optical schemes for viewing biological material with a microscope while it is being centrifuged. Several successful experiments have been carried out on the sedimentation of small particles and molecules from solution.[7, 15, 18, 19] A few successful attempts have been reported where the apparatus has been used for the determination of molecular weights.[15, 18, 19] In these experiments it is necessary to take elaborate precautions to maintain temperature equilibrium throughout the centrifuge "bowl" in order to prevent "remixing" or stirring.[8, 18, 20] Qualitatively it is easy to see that this is essential because the principal factor which produces movements in a liquid is roughly proportional to the "differences in density" multiplied by the centripetal acceleration. Hence stirring may result, even in a pure liquid, if inequalities in density arising from nonuniformity of temperature exist because of the large value of the centripetal acceleration. As a matter of fact this factor is so great that, in an ultracentrifuge, it would be practically impossible to prevent stirring if it were not for a second factor which tends to counterbalance it. This arises from the fact that

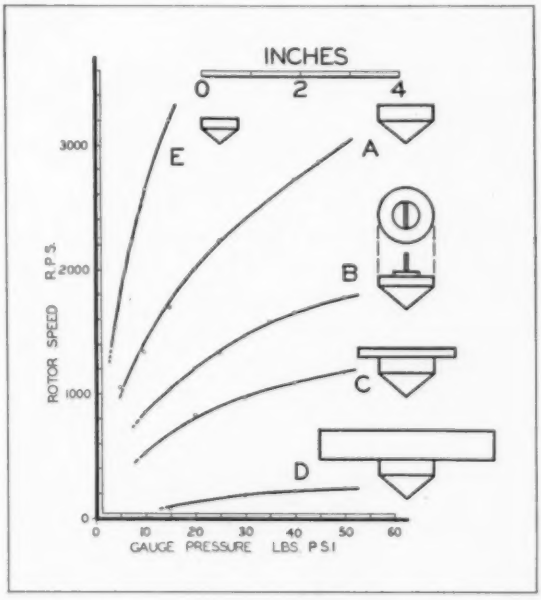

FIG. 5. Curves of the relation of air pressure to rotor speed for the case where the air surrounding the rotor is at atmospheric pressure. Curves *A*, *B*, *C*, and *D* were taken with the same stator (Figs. 1 and 2) and the rotors sketched to scale at right of each curve. Curve *E* was taken with stator shown in Fig. 3. For curves *A*, *B*, *C*, and *D* with driving pressure of 15 lb. per in.^{2}, 4.4 cu. ft. of air per minute was required. For curve *C* under same conditions, 5 cu. ft. per min. was required.

FIG. 6. Image of 8 mm bismuth spark swept from left to right across the photographic plate perpendicular to axis of electrodes by a rotating mirror mounted on an air turbine. Note that the intense luminosity due to air, initially reaching across the gap, soon dies out and that the luminosity from the metal vapor initially starts from each electrode and progresses slowly toward the opposite one. Complete picture shown represents about 4×10^{-6} sec.

the density of a liquid increases with the pressure. Since the density of the liquid increases toward the periphery, due to increased pressure, stirring caused by temperature gradients is counteracted to a certain extent.*

It is especially difficult to maintain temperature equilibrium in the simple air-driven centrifuge which spins in air at atmospheric pressure. First, as can be seen from the curves of Fig. 5, the air friction is so great that the moving surfaces must be considerably heated. This heating is greatest near the periphery and top surface but zero at the center so that the temperature gradients are in the direction to produce stirring in most practical cases. Second, the expanding air jets striking the under side of the rotor may cool it several degrees and hence tend to produce additional temperature gradients from bottom to top of the centrifuge. This difficulty of maintaining temperature equilibrium in the above type of centrifuge, the increased power necessary to drive large rotors in gases at atmospheric pressure and the general desirability of spinning rotors at very high speeds in a vacuum, stimulated us at Virginia to devise the vacuum-type ultracentrifuge.

Air-Driven Vacuum Type Ultracentrifuge

In the original vacuum type air-driven ultracentrifuge[21, 8] the large rotor or "centrifuge" inside a vacuum chamber was driven and supported by an air turbine such as shown in Figs. 1 and 2 located outside and vertically above the chamber. The turbine and centrifuge were connected by a flexible shaft (steel piano wire) which was coaxial with their axes of rotation and passed through a vacuum tight oil gland or bearing that sealed the vacuum chamber. With this arrangement the centrifuge could seek its own axis of rotation (within limits) and hence was very stable. Also the vacuum attainable depended upon only the vapor pressure of the oil which could be made very small (10^{-6} to 10^{-7} mm for some oils). Furthermore, since the efficiency of the air-driven turbine was not appreciably decreased by the added weight of the centrifuge or by the bearing friction the maximum rotational speed was set only by the mechanical strength of the centrifuge. In the type of air turbine shown in Fig. 1 the turbine is both supported and driven by the same air jets. This naturally gave trouble in bringing the centrifuge to rest because when the

FIG. 7. Method of spinning rods or tubes in air at atmospheric pressure. Stators at top and bottom are flexibly mounted and damped.

* The maximum stable or adiabatic radial temperature gradient for a substance in a centrifugal field can be calculated from the familiar thermodynamic equations. $dT/dp=(T/c_p)(\partial v/\partial T)_p$ where the symbols have the usual meaning. For air at 20°C treated as an ideal gas we have, approximately $dT_r/dr=(d/dr)(5\times10^{-8})w^2r^2=10^{-7}w^2r$. Where the field is a million times gravity $dT_r/dr=100$ deg./cm app. In the case of water at 20°C $dT/dp=1.8\times10^{-9}$ deg./dyne·cm². Since $dp/dr=w^2r$, $dT_r/dr=1.8\times10^{-9}w^2r$. For a million times gravity $dT_r/dr=1.8$ deg./cm app. The above calculations were taken from Professor L. G. Hoxton's lectures in thermodynamics.

driving air pressure was reduced, unless considerable care was taken, the turbine often "grabbed" and broke the flexible wire. Accordingly a driving turbine was constructed[8] in which the cushion of air that supported it was independent of the air jets that spun it. This made it possible to turn off the driving air and allow the centrifuge to coast smoothly to rest on the air cushion support. Several different workers have adapted and improved the above vacuum-type centrifuge to fit their special problems,[22–32] but unfortunately space does not permit a detailed review of these. Instead a brief description will be given of the essential features and of apparatus which has been used successfully at the University of Virginia for different purposes.

Figures 8 and 9 show a drawing and picture of an apparatus used for the separation of isotopes by evaporative centrifuging,[31, 32] while Figs. 10 and 11 show an arrangement constructed for research in biochemistry. The rotating members consist of the driving turbine, T, the flexible shaft, A, and the centrifuge or large rotor, C. The flexible shaft, A, passes through two vacuum tight oil glands (or bearings), G_1 and G_2. G_1 seals the vacuum chamber. G_2 is added so that C may be filled with liquid or gas or evacuated while spinning. G_2 also permits the use of the very stable "flat type" of air bearing support. G_1 and G_2 are constructed by machining a brass rod to the shape shown in Figs. 8, 10 and 12, and by boring a hole (usually about $\frac{3}{8}''$ diameter) along its axis. A plug of bearing metal (about $\frac{1}{4}''$ to $\frac{3}{8}''$ long) is then fitted into each end of this hole and another hole bored along the axis of each plug slightly smaller in diameter than the shaft A. These holes in the plugs are then reamed to such a size that A just slips in without forcing. The kind of bearing metal is determined by the material of A. When the latter is steel the former may be brass, bronze, babbitt metal, etc. G_1 and G_2 are mounted, as shown in the figures, in round flexible rings (made of Duprene[33] or other flexible oil-resistant materials). Vacuum pump oil is forced into G_1 and G_2 through the channels shown in Figs. 8

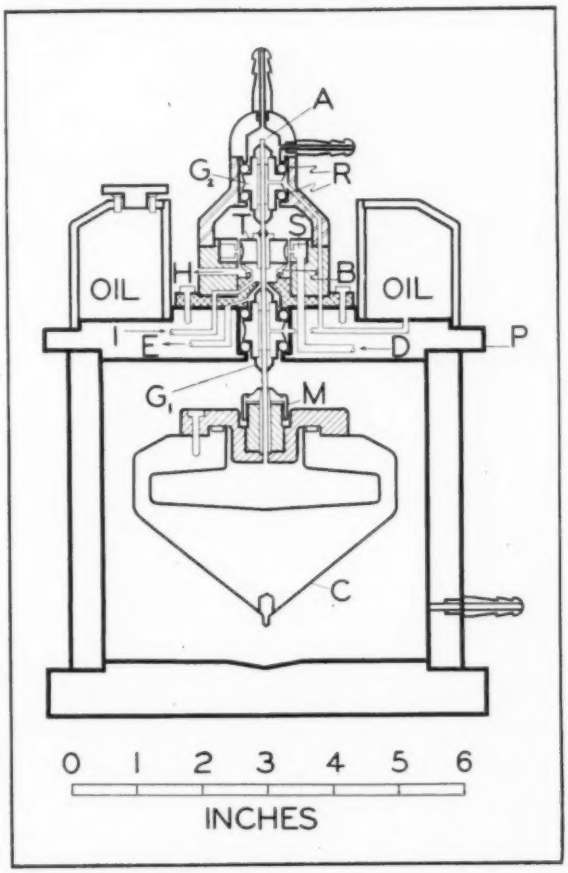

FIG. 8. Drawing approximately to scale of air-driven vacuum type centrifuge employed in the concentration of isotopes.

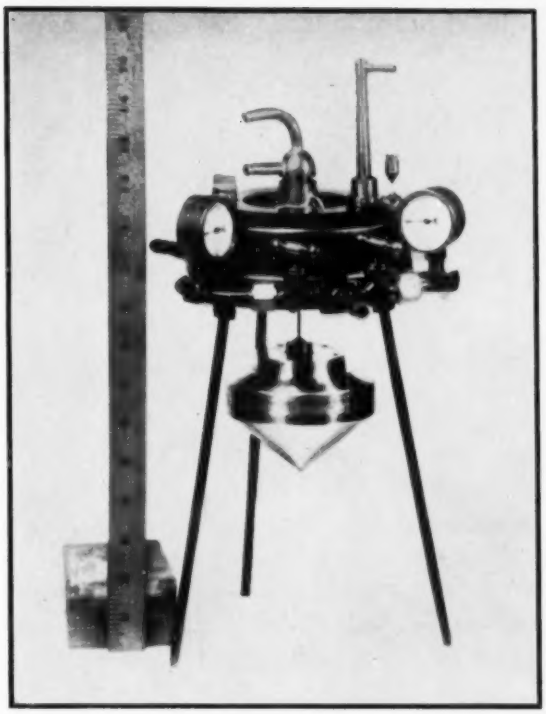

FIG. 9a. Photograph of apparatus of Fig. 8 with vacuum chamber removed. Inch scale on left.

and 10. If properly made, less than one cc of oil will leak into the vacuum chamber per hour through G_1. This type of mounting, besides giving the proper flexibility to G_1, allows any heat generated in the bearing to be carried out by the oil to the plate P. That is, the shaft A is held at approximately the same temperature as the vacuum chamber which, in turn, may be thermostated easily by standard procedure. The flexible shaft A may be either a rod or a tube. A rod is usually preferable when it is not necessary to fill or evacuate C while spinning, although stainless steel hypodermic needle tubing serves almost as well.[34] The diameter of A depends upon the weight and diameter of C. For the $4\frac{1}{4}''$ Duralumin rotor shown in Figs. 8 and 9 hypodermic needle tubing of No. 15 Stubbs gauge is used, while for the rotor, Figs. 10 and 11, the diameter of A was gauge 14. Besides steel and stainless steel, A may be made of any reasonably strong material such as phosphor-bronze, beryllium-copper or German silver. The latter is especially useful when C is spun at low temperatures (dry ice or liquid air) while some of the others are useful in magnetic experiments. The diameter of the "pill box" shaped turbine, T, is also determined by the weight and speed of C. In Figs. 8 and 10 the diameters of T are $\frac{3}{4}''$ and $\frac{7}{8}''$, respectively. The turbine should be made of strong material such as Duralumin, phosphor-bronze or steel. The type of fluting is clear from Figs. 9b and 12. Preferably the under surface of T should be flat when an upper bearing G_2 is used but cone-shaped (apex angle 90° to 92°) when the upper bearing is omitted. The air cushion which supports the turbine is formed between the slightly cone-shaped collar, B, and the under surface of T. B should preferably be made of Bakelite although Duralumin is satisfactory. B rests upon a flexible round or flat ring (Duprene or rubber). The stator, S, is a simple air box with channels bored to direct the driving air jets. The channels have their axes in a horizontal plane and if extended would pass within about $\frac{9}{32}''$ and $\frac{11}{32}''$ of the axis of the turbine in Figs. 10 and 12, respectively. The number and size of the channels depend upon the load and size of T. For Figs. 8 and 10, each has 8 channels bored with a No. 60 twist drill. It should be noted that if the number of flutings on the rotor is odd, the number of channels in the stator should be even and *vice versa*. C is always specially designed for the problem under investigation. A pin or screw clutch, M, fastens A to C in such a way that a lead or soft metal washer between the two makes the connection vacuum tight. A few of the different shapes and sizes are shown in Fig. 13. Special attention should be given to the material and shape of C because its bursting strength is the only factor that limits the rotational speed attainable. Fig. 14 shows a few rotors after they have been exploded by their own centrifugal force as well as some of the vacuum chambers which surrounded them. For solid rotors the bursting speed is set not by the absolute strength of the material but by the absolute strength divided by the density. However, in centrifuge work this is often no longer true and the absolute strength becomes important. At the present time among the lighter alloys Duralumin ST14 (forged) is probably the best general purpose material although in some cases special steels are superior. A large amount of both theoretical and experimental work has been done on the materials and shapes of rotors to give them maximum bursting speed. Nevertheless materials that

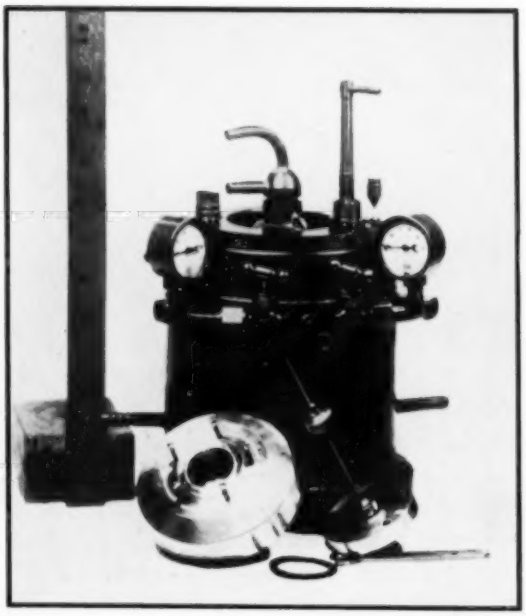

FIG. 9b. The same as 9a with vacuum chamber in place. Rotor parts together with flexible shaft and turbine are in foreground.

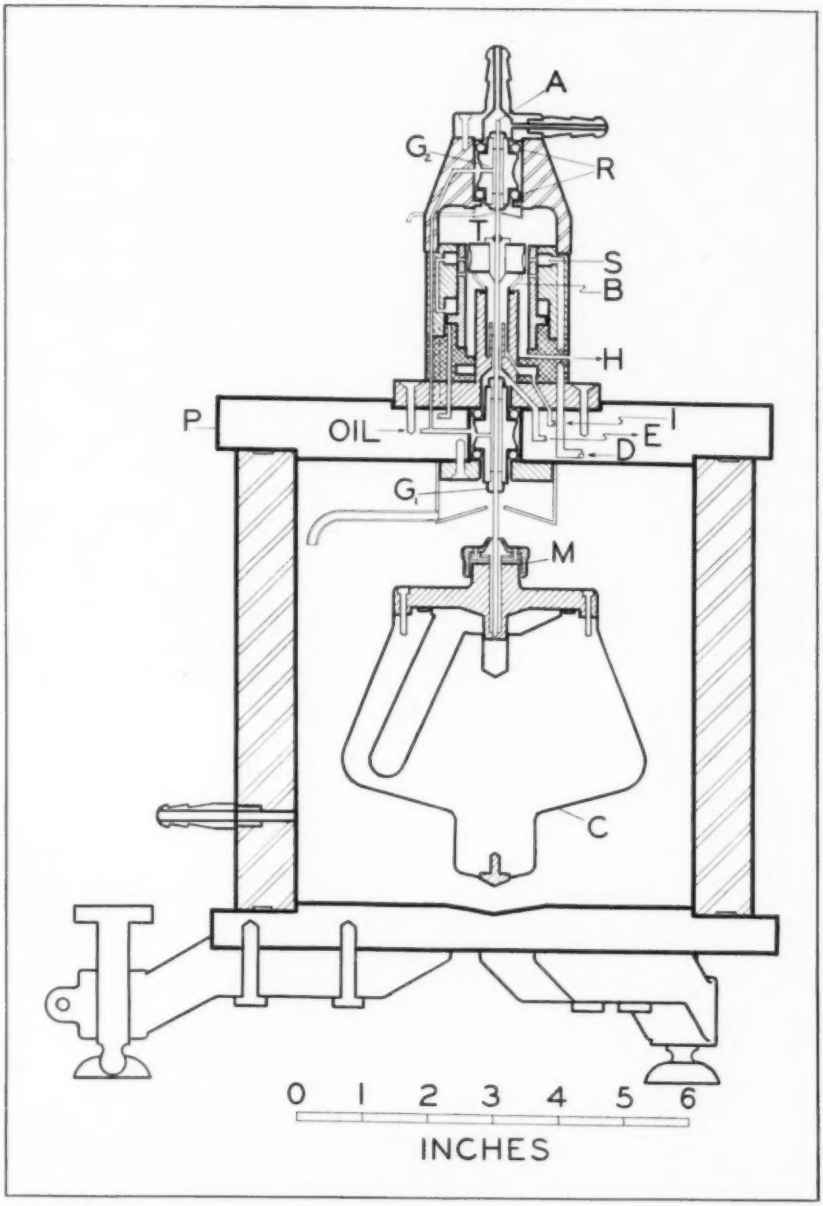

FIG. 10. Drawing approximately to scale of air-driven vacuum type centrifuge made at Virginia for Dr. Alfred Chanutin for research in biochemistry.

will stand higher rotational speeds are at present the greatest need in almost every problem undertaken with the ultracentrifuge. It might also be noted that in a rough way the strength of a homogeneous rotor depends upon the peripheral velocity squared, or w^2r^2, where w=angular velocity and r=radius, while the centripetal acceleration is given by w^2r. Hence when it is desired to obtain large accelerations by the above method small rotors should be employed.

At the time of explosion the peripheral speed of one of the rotors shown in Fig. 14 was 8×10^4 cm/sec. or about 2.6 times the velocity of sound. Hence the vacuum chamber must be either heavily barricaded or made very strong to protect the operator in case of an explosion. The latter procedure is preferable although the former is less expensive. Bauer and Pickels[20, 22] recommend that the vacuum chamber be a heat treated nickel-steel alloy cylinder with wall thickness of at least an inch for the type of Duralumin rotors used in their work, while Pollock and Collie[35] employ a short segment of a discarded cannon barrel for their chamber. In both cases, the tops and bottoms of the chamber should be steel plates at least an inch in thickness. For many purposes the temperature of the chamber must be carefully controlled. This can be done by forcing a liquid at the proper temperature through copper pipes in good thermal contact with both cylinders and end pieces.

To operate the apparatus, C is clamped at the desired position on A, usually found by trial ($\frac{3}{8}''$ to $\frac{3}{4}''$ from lower bearing is satisfactory in Figs. 8 and 10 although longer distance may be used if specially desired). Air is then admitted through the channel, I, and the pressure increased until the rotating parts are freely supported upon the air cushion formed between B and T. (This pressure was about $6\frac{1}{2}$ lb./in.^{2} and $9\frac{1}{2}$ lb./in.^{2} for the instruments shown in Figs. 8 and 10.) Air pressure equal to or slightly greater than that

FIG. 11a. Photograph of apparatus of Fig. 10 with vacuum chamber removed.

required to maintain the above air cushion is next applied to the oil tank which forces the vacuum pump oil into the glands G_1 and G_2. The apparatus is then placed in its position upon the vacuum chamber and the vacuum pumps are started. A convenient pumping arrangement consists of an oil diffusion pump backed by, say, a Cenco Hyvac pump. If C must remain at constant temperature the pressure should be below one micron. The driving air is then admitted through the channel, D, to the stator, S. This starts the centrifuge spinning. As the rotors accelerate, they pass through certain speeds at which vibration takes place. Fortunately, these are not violent and consist of rather narrow bands. The first occurs when the rotational frequency approximately equals the frequency with which C will vibrate if, while not spinning, it is given a displacement and allowed to "vibrate as a pendulum." Other critical vibration frequencies due to resonance points in the apparatus, natural vibrations in C itself, standing waves in A, etc., occur at higher rotational speeds. These have harmonics at still higher frequencies, but their amplitudes are very small. It is good practice to carry the centrifuge through the critical speeds as quickly as possible. Also a working speed should be selected which is well removed from a vibration frequency regardless of its amplitude. Sometimes when the apparatus is over-cushioned or over-damped it is difficult to detect these minute vibrations. Hence no more flexibility and damping should be given to the driving parts than necessary. Except at the few critical speeds discussed above, the centrifuge spins extremely steadily and smoothly. Even when examined with a microscope or by reflected light at a distance of a meter the centrifuge shows no observable wobble or vibration. As a matter of fact if properly adjusted the centrifuge accelerates so smoothly that a careful watch must be kept upon its speed to prevent exceeding its "bursting speed." With the apparatus of Figs. 8 and 9 the centrifuge is accelerated with 35 to 40 lb./in.^{2} driving air pressure. It is maintained at working speed (1550 r.p.s.) by 10 lb./in.^{2} driving air pressure. (The volume of air at 10 lb./in.^{2} is 2 ft.^{3}/min. N.T.P.) If the above centrifuge should reach 1650 r.p.s. (found experimentally) it would

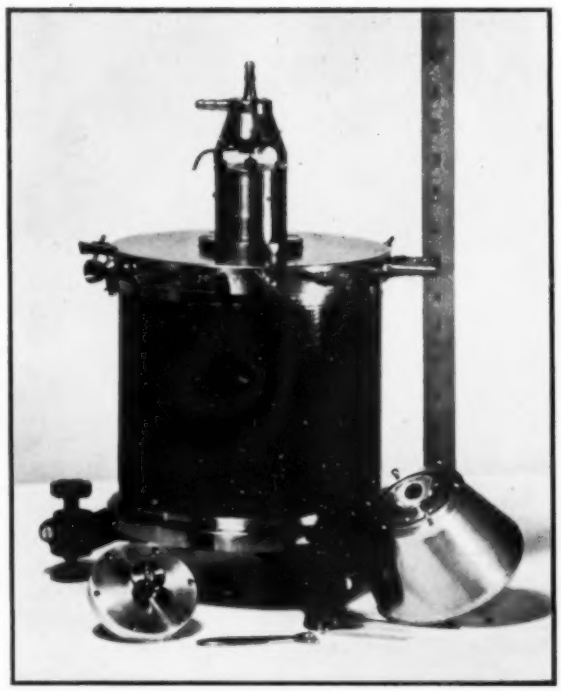

FIG. 11b. Same as 11a with vacuum chamber in place and centrifuge parts in foreground.

start to deform. This emphasizes the fact that it is necessary to maintain not only a constant driving air pressure but an almost continuous check upon the rotational speed. Usually reducing valves and air strainers of various kinds are used to keep the air clean and at constant pressure, while the stroboscopic method is used to measure the speed. Several methods have been described with which it is possible not only to

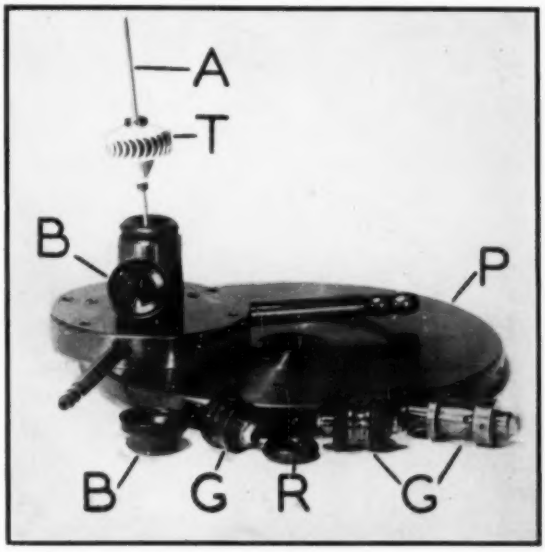

FIG. 12. Photograph of turbine, flexible shaft, Bakelite collar, round Duprene rings, vacuum tight oil glands, and top plate of vacuum chamber. The lettering corresponds to that in Figs. 8 and 10.

measure the speed but to regulate it as well.[3, 36–38] In the apparatus of Fig. 9, if C is allowed to coast in coming to rest from 1500 r.p.s. it will require more than a day unless air is admitted to the vacuum chamber. For this reason reverse jets are provided in the apparatus of Fig. 10 so that the centrifuge may be stopped smoothly in a few minutes. With an apparatus similar to that shown in Fig. 9 one set of plugs (babbitt bearings) in the glands G_1 and G_2 usually last, if care is taken in starting, until the rotor has made more than 10^9 rev.

Inverted Ultracentrifuges

In many experiments it is necessary to place observing apparatus directly above and on the axis of the centrifuge. Obviously this is impossible with the apparatus described above because

FIG. 13. Photograph of a collection of rotors that have been spun in a vacuum.

of the location of the driving turbine. In order to overcome this difficulty the apparatus shown in Fig. 15 was devised[39] in which the driving turbine, air cushion, supports, etc., are placed below rather than above the vacuum chamber. In this arrangement the centrifuge, instead of hanging on a flexible shaft supported by the turbine above the vacuum chamber, is mounted on a flexible shaft extending upward through a vacuum gland and supported by a turbine below the chamber. The driving and supporting mechanisms are essentially the same as those described above. In practice the stability of this inverted type of centrifuge is as great as if not greater than that of the type (Figs. 8–12) previously described. Like a top spinning on its point any gravitational couple produces a precession which is in the direction of rotation rather than opposed to it as in the original type.

Long Rods and Tubes

In the course of some experiments at Virginia we found it necessary to develop the technique of spinning long vertical rods and tubes in a vacuum. They are supported by flexible shafts, one above and one below the rotor. These shafts, which may be either tubes or rods, pass through oil glands similar to those described previously in the top and bottom of the vacuum chamber respectively. The spinning rod or tube is supported and driven by the type of air cushion and turbine already described in connection with Figs. 8–12. This driving mechanism may be either above or

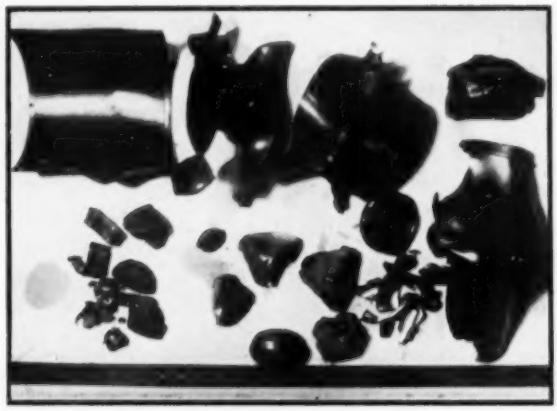

FIG. 14. Photograph of metal rotors and vacuum chambers after explosion. At right are parts of an exploded steel tube.

below the vacuum chamber. It is generally necessary to make the length of shaft between gland and rotor greater than usual. Also, it is desirable to make the mounting of the glands more flexible. However, these adjustments depend upon the length, diameter and weight of the rod or tube. Rods 2.5 feet long and weighing 75 lb. have been spun. Again, the only factor which determines the maximum rotational speed attainable is the bursting strength of the tube or rod. Fig. 14 shows a steel tube 1 foot long and 2.6 inches in diameter exploded by Mr. C. Skarstrom, at Virginia.

Uses of Air-Driven Vacuum Type Ultracentrifuges

Perhaps the most extensive use of the air-driven vacuum type centrifuge has been in the concentration and purification of biological materials. The pioneering work of Bauer and Pickels[20, 22] on the concentration of yellow fever virus, of Wyckoff and his collaborators [23–30] in their investigations of so-called "macromolecular" proteins and the experiments of Severinghaus and Chiles on the concentration of hormones illustrate the possibilities of the ultracentrifuge in biology, medicine, and chemistry. In addition to the above, the air-driven vacuum type centrifuge has been employed in the determination of molecular weights by the classical methods of Svedberg.[20, 22–30] In this work it has been found to yield as accurate results as Svedberg's oil turbine ultracentrifuge.[20, 24] In connection with the above types of work there is need for additional theoretical work on the problems of sedimentation in both liquids and gases (with accompanying tables, etc.). Something similar to the excellent work of Mason and Weaver[40, 41] for settling in liquids in uniform gravitational fields would be exceedingly useful. The vacuum type of ultracentrifuge has also been utilized for the separation of gases and for velocity selection of molecules.[31] As mentioned previously, the apparatus in Figs. 8 and 9 is now being employed in the separation of isotopes.[32] The above apparatus may also be used to study the strength of materials, as an alternator, for mechanically making or breaking electrical circuits, to spin rotors at either high or very low temperature, to spin mirrors at high speeds, to spin rods or other rotors for the study of gyromagnetic phenomena, for moving two surfaces past each other at very high speeds, for rotating drums, and in general in the study of phenomena which occur in intense centrifugal fields.

Electrical, Steam and Oil-Driven Ultracentrifuges

In many laboratories compressed air in sufficient quantities to operate the air-driven ultracentrifuge is not available. Also there is the need for an ultracentrifuge which is comparatively "fool proof" and requires a minimum of attention of the operator, i.e., one which automatically speeds up to the required angular velocity and remains at this value until changed by the operator. Recently an electrically-driven vacuum type ultracentrifuge[42] has been described which has promise of satisfying the above needs. In this apparatus a small electrical motor is used to drive the centrifuge in place of the air turbine while the air cushion support is essentially the same as that previously described (Figs. 8–12). This is a great saving of air because the amount used in the cushion support is small in comparison with that required to drive the turbine. Also it need only be furnished at from 5 to 15 lb./in.^{2} Hence a small, relatively inexpensive air compressor is all that is required. The armature of the electrical motor must be specially designed to stand high centrifugal forces. Both induction and synchronous motors have been used in connection with variable frequency thyratron inverter

circuits. Variable frequency oscillator circuits employing high vacuum tubes or other types of alternators[43] also may be used to give the necessary a.c. power. As a matter of fact it is highly probable that a d.c. motor could be made to serve the purpose almost equally well. The electrically driven centrifuge has been used successfully up to 1000 r.p.s. and there seems to be no reason why this method, when fully developed, cannot be made to attain much higher rotational speeds.

The steam-driven vacuum type ultracentrifuge[44] is essentially the same as the air-driven type except that precautions must be taken to insulate thermally the driving turbine from the vacuum chamber. This has been accomplished by mounting the stator upon nonconducting material such as lavite and circulating water in copper coils in thermal contact with all of the upper part of the vacuum chamber. The steam is generated electrically (5 kw water heater unit) and is superheated. The only factor which determines the maximum speed attainable (as in the case of the air-driven centrifuge) is the bursting strength of the centrifuge.

The oil-turbine ultracentrifuge was developed by Professor Svedberg and his students[45–57] and used by them in their epoch-making experiments on the determination of the molecular weights of proteins, etc. The apparatus represents many years of development and is a model of machine design and precise workmanship. The centrifuge is supported in horizontal bearings and is spun by twin oil turbines, on each end of a large rigid shaft. The oil which drives the turbines is supplied by a special compressor and is cooled by a refrigeration plant to a suitable temperature before it enters the turbine chambers. The centrifuge spins in circulating hydrogen at about 25 mm pressure. The rotors are made of chromium-nickel steel. Svedberg was able to obtain his highest centrifugal forces with hard steel but discarded it in favor of softer steels which were found to be more reliable. The centrifuge or large rotor is shaped to give it maximum strength and must be dynamically balanced with extreme care. Reference should be made to the papers of Svedberg[45, 46] for a detailed description.

Recently a promising new method of producing high rotational speeds in a vacuum or in gases at various pressures has been made possible by the Holmes magnetic suspension.[58] This consists of a vertical rod of iron suspended in the coaxial field of one or more solenoids. A steady direct current in one solenoid produces a field which almost supports the iron rod to which are attached the other rotating parts. The rotor or vane attached to the rod controls the amount of light incident on a photo-cell. The current from this cell is amplified and fed to the solenoid in such a manner as to automatically maintain the rod at a predetermined height. Such a suspension is macroscopically stable and, in a vacuum, theoretically may have infinitesimal torque against axial rotation. In the preliminary experiments[58, 59] a small rotor (mass and moment of inertia of iron rod and attached Duralumin rotor was 6 g and 0.8 g cm^2, respectively) in a vacuum (10^{-5} mm) was spun to 1200 r.p.s. by the action of the field of a small magnet mounted horizontally on an air-driven turbine spinning below the rotor at 1500 r.p.s. Damping observations were carried out in the neighborhood of 600 r.p.s. with the driving magnet removed. The deceleration observed was 2×10^{-3} rev./sec.^{2} corresponding to a frictional torque of about 10^{-2} dyne cm. This low frictional torque makes possible the production of very constant rotational speeds. For example, if the rotor is driven by the action of a rotating field, the field need not be large and hence can be accurately controlled both in amplitude and frequency (say by a piezoelectric circuit). The large slip resulting from an effectively weak field makes the rotor speed fluctuate much less than that of

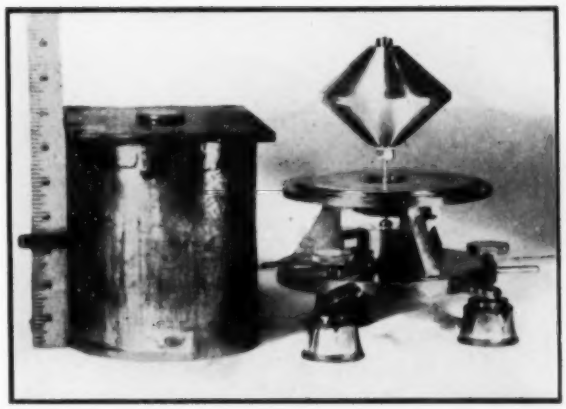

FIG. 15. Photograph of an air-driven inverted vacuum type ultracentrifuge with vacuum chamber removed.

the rotating field. The experiments indicate that it should be possible to spin rotors of almost any size, which can be surrounded by a vacuum chamber, up to their bursting speeds and that the method should be of considerable use in experiments where extremely high vacuum or constant rotational speeds are required.

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The Importance of Observations from the Upper Atmosphere in Long Range Weather Forecasting

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THE term "long range weather forecasting" means essentially the same thing, whether it is applied to unit periods expressed in weeks, months, years, decades, or centuries. The forecast is an estimate of the degree to which the *average* of temperature, rainfall, and possibly other meteorological elements will be above or below the normal value for each of the successive unit periods, or for a definite season. This is in contrast to the ordinary daily short range forecasts, which are intended to predict accurately the sequence of the individual changes of the elements. Probably such detailed forecasts will never be extended beyond a few days in range, because they require an accurate estimate of the movement and development of each element of disturbance in the pressure field, or of each air mass and frontal system. Air mass and frontal systems, together with corresponding disturbances of the pressure field, are in a continuous process of formation and disintegration, but it seems rather improbable that the detailed development of systems as yet unborn can ever be forecasted.

On the other hand, we know from observation that frequently pronounced anomalies in the meteorological elements may persist over considerable areas for weeks, months, and even years. Such persistent anomalies must represent the integrated effect of certain abnormal characteristics of the short period weather sequences during these periods. The methods which have been used in the attempt to forecast these persistent anomalies of the meteorological elements are nearly as numerous as the investigators who have studied them. However, these methods fall into two general classes, which may be referred to as the *statistical* and the *synoptic*, or a combination of the two. The statistical methods include the frequent analysis of meteorological data in the search for regular periods which may be used for forecasting, as well as limitless correlations of present and past weather, ocean conditions, solar activity, and even planetary configurations with the future weather. The weakness of all such purely statistical methods of forecasting lies in the fact that they represent empirical shortcuts which have no concern at all with physical causes. Only the most restricted and routine use of the available meteorological data is made. Needless to say the results obtained by these methods have been of quite negligible value in the long run.

The basis of the synoptic methods is the synoptic chart, or weather map. The maps which are to be used for purposes of long range weather forecasting should cover an extensive area, preferably not less than a hemisphere. They should be prepared daily, and carefully analyzed by the

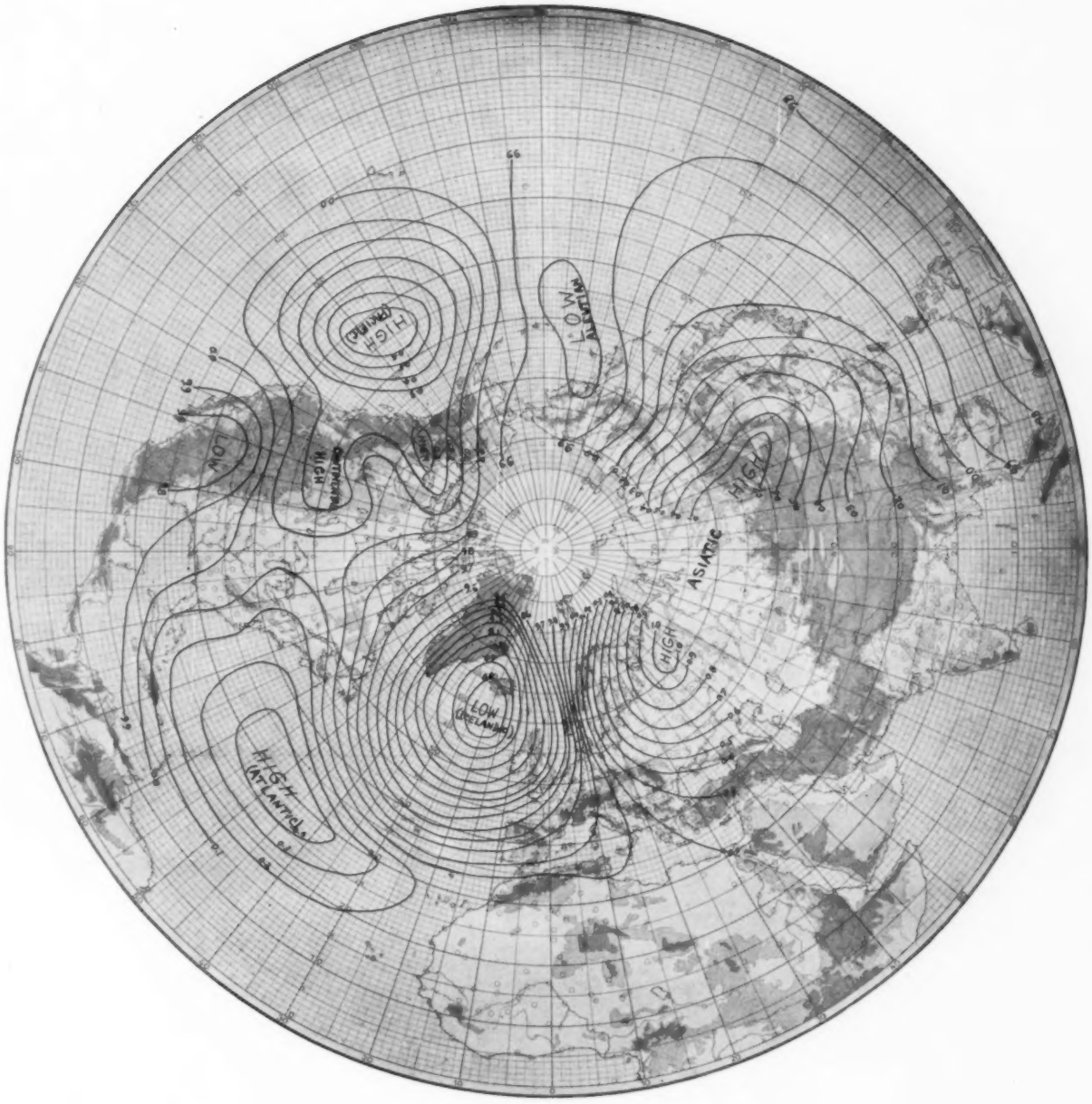
methods of air mass analysis. The preparation of such daily maps requires a large amount of routine work, a fact which probably explains why the great majority of the investigators of the long range forecast problem have shown marked preference for purely statistical methods of attack. On the other hand, the synoptic methods have the one great advantage, at the very outset, of furnishing an extensive survey, by means of the analyzed weather map, of those very atmospheric conditions whose continuance or change is to be forecast. Consequently the forecaster obtains a definite picture of the state of the general circulation on the earth's surface in relation to the particular meteorological elements and the particular area with which he is concerned. The statistical forecaster, on the other hand, forecasts by routine procedure the individual meteorological elements for a given area without any physical picture as to how these elements are interrelated in time or space. It should be a distinct advantage if the statistical investigators were to base their work on the synoptic map, or at least were to acquire a background of experience with such maps as a guide to the intelligent application of the statistical methods.

During the past year the careful analysis of a continuous series of northern hemisphere charts, once daily, has been carried on without interruption at the Massachusetts Institute of Technology. In this connection there have been prepared and verified regular weather forecasts for weekly periods one, two, and three weeks in advance for some ten stations scattered widely over the northern hemisphere. This undertaking is being supported in part by the Bureau of Agricultural Economics of the Department of Agriculture, as one phase of a general investigation of the long range forecast problem. Probably by outlining briefly certain aspects of the past year's work and by summarizing the principal conclusions which have been suggested by it, the significant features of the synoptic method of attack on the long range forecast problem can best be illustrated, and the line of attack which appears to offer most hope of further progress can be indicated. It is this further progress which will be dependent almost entirely upon adequate observations from the upper atmosphere.

During the past year careful analysis of a continuous series of synoptic charts of the northern hemisphere has been carried on at the Massachusetts Institute of Technology. In this paper, Professor Hurd C. Willett outlines the methods in use and summarizes the principal conclusions.

There were really two immediate more or less definite general aims behind this daily analysis of northern hemisphere weather maps at the Massachusetts Institute of Technology. These were (1) to acquire a practical acquaintance from day to day with the major features of the general circulation patterns of the northern hemisphere and the influence of the prevailing circulation pattern on the contemporary weather conditions over this hemisphere, and (2) to look for possible empirical clues as to the future state of the general circulation in its current state and present tendencies.

Concerning the first of these points, it has long been realized by all practical weather forecasters that there is a close correlation between the general circulation pattern and the location and intensity of all important areas of anomaly of the different meteorological elements. By the general circulation pattern is meant the location and intensity of the so-called centers of action, or the semi-permanent centers of high and low pressure of the northern hemisphere, with their corresponding systems of prevailing winds. The principal centers of action on the northern hemisphere during the colder half of the year are the Siberian and North American high pressure areas, the Icelandic and Aleutian low pressure areas, and the subtropical high pressure area over the Atlantic and that over the Pacific Ocean. According as these principal individual centers depart from their normal intensity and normal position during a given period, we have a corresponding departure from normal of the general circulation pattern, or of the mean air movement, over the northern hemisphere. Such a displacement and changed intensity of the major prevailing air currents results in displacements of the principal frontal zones, or zones of inter-



Map showing the development of the principal centers of action as indicated by the mean pressure field during the week of January 17–23, 1937 (the week of heaviest rainfall in the Ohio valley).

action between the polar and the tropical air currents. This means in other words that the zones of maximum storminess, or storm tracks, are shifted from their normal position, with resultant persistent anomalies in the fields of all the meteorological elements.

The experience gained with the northern hemisphere map during the past winter which was particularly rich in persistent anomalous conditions, illustrated perfectly the complete dominance of the weather conditions over large areas by the vagaries of the individual centers of action, together with the frontal zones of interaction between them, which go to make up the general circulation pattern (to be illustrated from slides showing mean pressure distributions). The unusual development and westward extension of the Atlantic high was evidenced by the abnormal

warmth in the eastern United States during the end of December and most of January, together with the extremely heavy rain in the Ohio valley along the frontal zone separating it from the cold continental anticyclone to the northwest. On the other hand the complete disappearance of the Atlantic center, and its replacement by abnormally low pressure from the region of Bermuda northward during the first week of February, brought the north Atlantic coast the only week of subnormal temperature during the winter months. An even more striking feature of the general circulation pattern last winter was the complete disappearance from the Aleutian Island region of the north Pacific of the usual Aleutian low, and its replacement by very persistent high pressure. This supplied the whole Pacific coast with abnormally cold polar Pacific air masses, which in turn were responsible for the formation of abnormally cold and persistent continental polar air masses over the western half of the continent. The usual mild maritime air of the north Pacific area moved northward to Bering Sea and western Alaska, instead of moving inland along the north Pacific coast. Consequently the precipitation was almost entirely snow instead of rain, and each storm was followed by an outbreak of very cold polar continental air. On the other hand, the Icelandic low was as persistently present and overdeveloped as the Aleutian low was absent. This was evidenced by an unusual persistence of severe storms over the north Atlantic and on the northwest coast of Europe.

Much more might be said about the complete dominance of the weather conditions over large areas during the past winter by the variations in the general circulation pattern, variations, as we have just seen, which depend upon the vagaries of the individual centers of action and of the frontal zones of interaction between them. If this winter can be taken as typical, it would appear quite definitely that the establishment of the general circulation pattern over the northern hemisphere is sufficient to determine the climatic characteristics of the corresponding period in most regions. It is scarcely necessary to point out, however, that such a correlation of the persistent meteorological anomalies with the general circulation pattern in no sense constitutes an explanation of the former, but that we are dealing rather with two manifestations of the same set of conditions. To offer an explanation of the condition, it is necessary to find the underlying physical causes of the variations in the details of the pattern of the principal centers of action of the general circulation.

With regard to the second immediate aim of the present investigation, that of finding in the existing pattern of the general circulation over the northern hemisphere clues to the coming changes or developments of importance in the circulation system, very little success has been met with up to the present. In other words, there seems to be offered little hope of finding any empirical basis in such circumpolar charts for the preparation of general weather forecasts one, two, or three weeks ahead, before the underlying physical causes of the anomalies in the general circulation are better understood. This result was, of course, anticipated, but it seemed desirable at least to make the effort. To aid in the search for such empirical indications, weekly mean pressure maps for the northern hemisphere were prepared regularly, as well as isallobaric maps for the weekly pressure changes. Such maps were found to be very helpful in bringing out the current trend in the development of the general circulation pattern, but such trends appear to be quite irregular in their persistence. No empirical indication of imminent changes in the current trend could be found on these maps.

Although the past winter's work has led to little progress in the direction of finding any empirical basis of long range weather forecasting by the synoptic method, at least it did serve to emphasize the close correspondence between the general circulation pattern and the persistence of anomalous meteorological conditions over large areas, and consequently to throw some light on certain aspects of the long range forecast problem. Certain conclusions were reached in the course of this year's work which indicate a further line of attack on this problem. These conclusions, however, are by no means final. They are not of the definite kind which can be conclusively proved, nor are they in considerable part original with the writer. They may be summed up about as follows:

(1) That there must be definite and persistent physical causes behind the more persistent

anomalous circulation patterns of the northern hemisphere. These patterns are far too irregular and persistent to be the result merely of chance interplay of the migratory cyclones and anticyclones of middle and northerly latitudes.

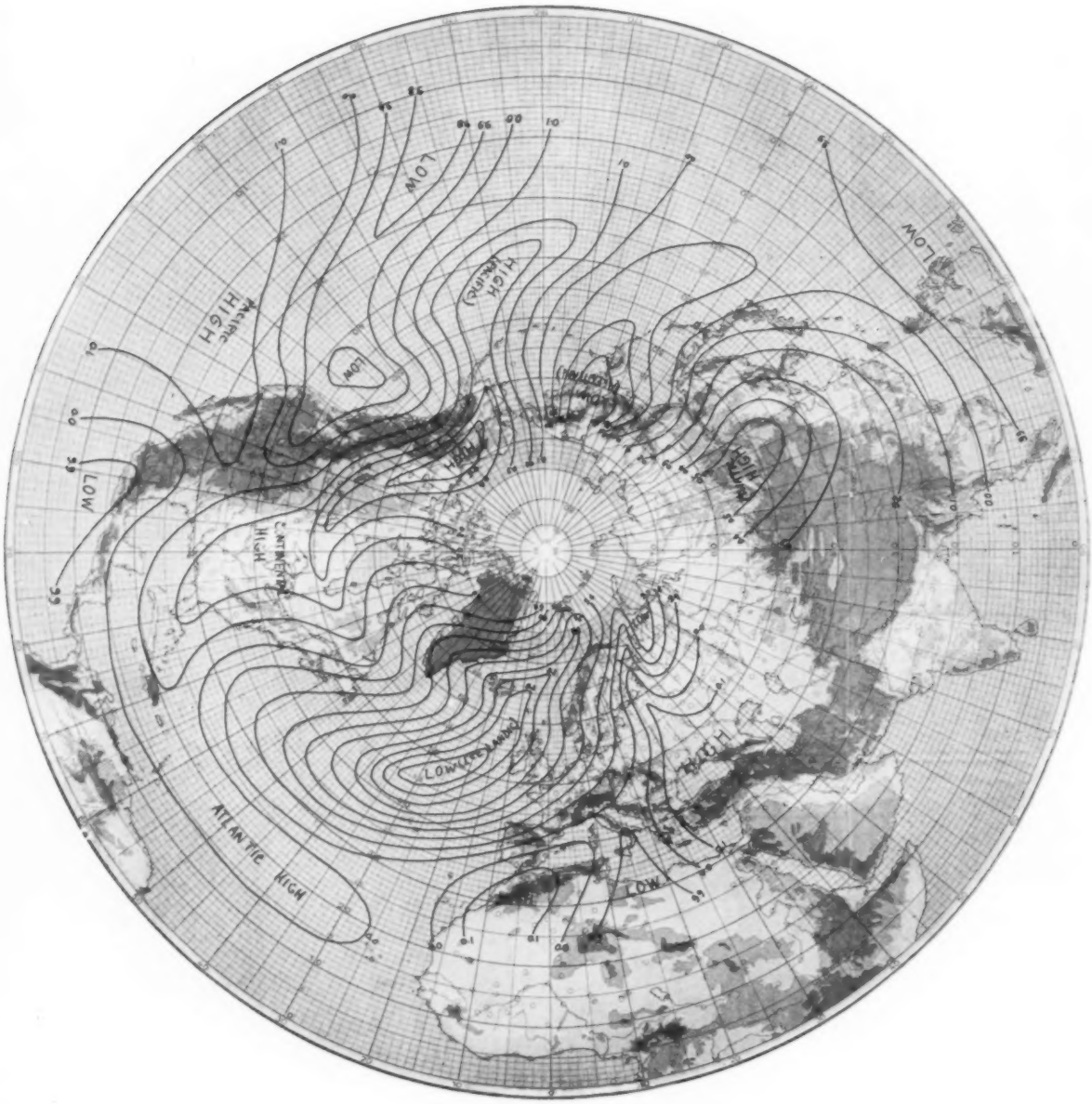
(2) That a real understanding of these underlying physical causes, and a quantitative knowledge of the different factors involved, will be necessary for even an approximate solution of the forecast problem. The complexity of conditions is such that empirical or statistical methods based on the direct observation of only one or two elements are bound to lead to conflicting and inconclusive results. This will be true even if the elements considered are actually the controlling factors, unless the mechanism by which they operate is understood. The preparation of the northern hemisphere charts performs the indispensable function of giving observational expression to the complex systems whose changes are to be forecast.

(3) That surface lag tendencies, notably the effects of short period glaciation or the irregular transport of ice and cold or warm water masses produced by temporary wind systems, are of very secondary if not negligible importance in the control of the general circulation pattern of the northern hemisphere, at least insofar as the fluctuations from week to week are concerned. This is evidenced by the abrupt and complete reversal of the prevailing conditions which is occasionally observed over large areas. Probably these surface lag effects play somewhat more of a role at high than at low latitudes. It is quite possible, however, that these lag effects may show in long period anomalies of small amplitude.

(4) That it seems to be the vast subtropical anticyclonic units in the general circulation pattern which are the prime movers in most important and persistent anomalous conditions of the general circulation. The cyclonic units at high latitudes, although completely dominating local conditions, appear to depend in both their location and their development on the behavior of the great primary subtropical units. Since surface conditions in the subtropics change comparatively little, this would seem to suggest that the essential factors of control are operative directly at upper levels in the atmosphere, whence the effects are recorded indirectly at the earth's surface in the general circulation pattern with its attendant distribution of the meteorological elements.

There are two general facts which become fairly evident from the discussion up to this point, namely (1) that persistent anomalies of the meteorological elements over large areas are directly connected with persistent anomalous patterns of the general circulation, and (2) that empirical or statistical methods are quite inadequate as a basis of forecasting changes in the general circulation pattern. Evidently, then, the line of attack which seems to offer most hope of further progress in the long range forecast problem must be based on the investigation of the mechanism by which a given circulation pattern is established, maintained, and terminated. The necessary first step in any such investigation, however, is to extend our present two dimensional northern hemisphere weather maps in the third dimension. The map of surface conditions over the northern hemisphere can now be made reasonably complete. At present there are still large areas from which few data are received, but this is caused rather by insufficient dissemination of the information than by the lack of observations themselves. Furthermore this condition is being improved rapidly. Probably within a year or two all the surface data desirable for the northern hemisphere map will be available, except perhaps from within the latitude circle of 75° N. On the southern hemisphere such surface information probably will be quite inadequate for a circumpolar map for many years to come.

But when it comes to a consideration of the upper atmosphere the available information, even on the northern hemisphere, is utterly inadequate. In order to complete our knowledge of the vertical structure of the general circulation we need the regular pilot balloon observations of wind direction and velocity, and the meteorograph records of pressure, temperature, and moisture aloft, from a widely scattered network of stations over the northern hemisphere. These observations should extend at least into the lower portion of the stratosphere. Only then will we have any clear conception of the complete state of the atmosphere associated with a definite surface pattern of the general circulation. At present we have only the most indefinite picture



Map showing the development of the principal centers of action as indicated by the mean pressure field during the week of January 31–February 6, 1937 (the week of coldest weather on the Atlantic coast of the United States).

even of the vertical extent and the level of maximum intensity of the individual centers of action, to say nothing of the changes which take place at high levels in connection with the major changes in the surface circulation pattern. There is no doubt but that the high level changes are frequently more extreme than the surface changes, and unexpectedly different from them. This involves the whole vital question of the interaction of the troposphere and stratosphere in atmospheric changes. About this interaction just enough is known at present so that its full significance is coming to be generally appreciated.

At the present time regular aerological observations are carried out only over the more civilized continental areas of the northern hemisphere, a restriction which gives practically no high level information whatsoever from the important centers of action. Furthermore the upper limit of much the greater part of this in-

formation which is obtained by small pilot balloons and aeroplanes, is between five and six kilometers, far short of the stratosphere heights which are needed. In addition, the dissemination of observations of this type is much more restricted than that of the ordinary surface observations. Consequently the synoptic worker is lucky to have available for his current map even from his own continent such aerological observations as are now being made. But in spite of the utter inadequacy of available aerological data at present, it is already proving invaluable in aiding our understanding of many details of the mechanism of the lower atmospheric circulation. However, a great advance in the amount of meteorological information available from the upper atmosphere should be realized during the next few years with the general introduction of the radio sounding balloon.

Until quite recently the only means of obtaining observations of pressure, temperature, and humidity aloft sufficiently promptly for use in current weather forecasting was by aeroplane. But aeroplane soundings could not be made in thick weather when they are most needed, nor could they be extended in general into the stratosphere. The only method of obtaining information from the stratosphere was by means of the ordinary sounding balloon, a method which is usually attended by a delay of weeks or months before the meteorograph is returned with its information, and one capable of use only in regions sufficiently inhabited so that the instrument stands a good chance of being found. Obviously such information was available only from restricted areas, and only after long delay. Consequently it was useless for current map work and forecasting, but could serve only for the historical study of past conditions in restricted localities. Within the past year or two the radio sounding balloon has been developed to such a point that these instruments can now be produced at a cost comparable to that of a single aeroplane sounding, and the reliability of the record is comparable to that of the aeroplane meteorograph. But this instrument has the further great advantages that its record is received immediately at the ground station, that it may be released and the record obtained at any point on land or sea in all kinds of weather, and that it may be counted upon to penetrate well into the stratosphere. There is no reason why within a few years there should not be a well-distributed network of a hundred or more such stations over the northern hemisphere, furnishing regularly from all regions exactly that aerological data which is so much needed. Direct observations of high level winds would be relatively unnecessary if the pressure distribution at such levels were accurately determinable from radio sounding balloon observations. However, direct observations of wind would in any case furnish a desirable check, and a greater detail of information.

The second step necessary to advance the general line of attack on the long range forecast problem as suggested here, involves the acquisition of all that observational information of a type not now included in routine meteorological measurements, but which doubtless will be essential to a further understanding of the physical causes which establish and terminate anomalous patterns of the general circulation. If it is assumed that the meteorological observations available are sufficient so that the major features of the existing thermal and dynamic state of the atmosphere are reasonably well known, then a successful long range forecast must depend upon our understanding of the factors which control the activity of the different major centers of action of the existing circulation pattern. So little is known at present about these controlling factors that progress in this direction will doubtless be very slow. It is impossible as yet to be definite even in suggesting an observational program. Such a program must be adapted to the indications which are obtained as the various possibilities are tested theoretically and observationally. Statistical correlations and periodicities may be helpful in this connection in indicating probable significant factors.

Undoubtedly the general problem of the heat balance in the atmosphere, in its many and complicated phases, is the basic problem which must be solved. In the last analysis not only the general circulation as a whole, but even the individual centers of action in this circulation depend upon localized unequal heating and cooling of the atmosphere. This suggests at once a number of factors which must be given quantitative consideration, both theoretical and observational.

In the first place must be considered variations of the effective insolational energy. Changes in the solar constant, especially those associated with the variations of sunspots and other evidences of solar activity, have long been indicated statistically as playing a role in long period weather changes. But our understanding of the physical mechanism by which these influences act consists of only the vaguest hypothesis. Certainly careful measurements not only of the solar constant, but especially of the energy output of the sun in the ultraviolet portion of the spectrum, should be systematically recorded. This type of measurement calls for mountain observatories. The intensity of the ultraviolet radiation is especially significant for our purpose owing to its effectiveness in producing ozone. Variations of the ozone content of the atmosphere at high levels, variations which are known to be large, should be systematically determined, and their quantitative effects on the absorption and return of outgoing long wave radiation should be carried out above the principal water vapor strata and cloud layers of the atmosphere. The effect of widely varying amounts of volcanic ash in the upper atmosphere in reducing the incoming solar radiation might well be checked, especially insofar as this reduction is the result of selective scattering. Finally, in the heat balance problem, there are the very important thermal effects produced in the atmosphere by the nature of the ground surface beneath. Ground surface influences vary greatly according as the surface is continental, oceanic, or glaciated. Such influences depend upon differences in albedo, in heat capacity, in radiative characteristics, and in surface roughness. These influences are transmitted to the overlying atmosphere by conduction, by direct radiation, and by the mechanical turbulence produced by the surface friction. Variations in the underlying surface in a given region, such as those between summer heating and winter glaciation of continental areas at high latitudes, or possibly also at times such as those due to the variation of the heat transport in ocean currents, may produce a corresponding profound modification of the general circulation pattern in the atmosphere.

It should be mentioned perhaps in concluding that mechanical as well as thermal influences must be considered in their influence on the general circulation pattern. However, it seems probable that the mechanical influences are important only in the short period fluctuations of the circulation pattern which are responsible for the day to day weather sequences. That problem might be treated by the methods of fluid mechanics. But the persistent anomalies of the general circulation with which we are concerned in long range weather forecasting are doubtless essentially thermal in origin. At the same time it seems evident that there must be a permanent mechanical effect due to differences of surface friction over continents and oceans which must find expression in the mean circulation pattern.

It is pointless to speculate any further at present as to particular factors which may control the general circulation pattern, or as to the particular observations from the upper atmosphere which are needed for their study. The extreme complexity of the problem is evident. Nevertheless, the very fact that definite anomalous patterns of the general circulation are observed to persist for long periods of time is in itself an indication that there must be some system and persistence to a given set of control conditions. This encourages the belief that it should eventually be possible to gain sufficient understanding of the controlling factors so that such persistent anomalous meteorological conditions may be reliably forecast.

Electric Breakdown of Solid and Liquid Insulators

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IN starting a discussion about electric breakdown in gases we have with others at least some ideas in common. The importance of impact ionization by electrons; the cumulative effect of successive impact ionization resulting in Townsend avalanches of electrons and positive ions; the necessity to make the discharge independent by regenerating the electrons needed for its maintenance. Such a background of universally acknowledged ideas is missing in the field of solid and liquid insulators in spite of the great and most valuable material offered by the research of the industrial and physical laboratories. The uncertainty of the situation, covered by such conceptions as "heat breakdown" and "electric breakdown" becomes evident as soon as any theoretical prediction is demanded.

A natural way out of this situation would be to go back to the knowledge gained in the field of the gas discharges and to consider step by step how the atomphysical aspect changes when we proceed to the condensed phases. The paper here presented aims to be a contribution to this line.

1. Paschen's Law and the Failure of Its Extrapolation

Paschen[1] found that the breakdown voltage of a spark gap tested under the gas pressure p with a gap distance d depends only on the product $p \cdot d$ (Fig. 1). Since the breakdown occurs by the ionizing impacts of electrons, the mean free path λ of these electrons is the adequate atomphysical standard for describing the observation. The gas pressure p is inversely proportional to λ; hence Paschen's statement is the expression of a similarity law. The breakdown voltage depends on the number d/λ of electron impacts across the gap. Tacitly the summing up of distances λ implies the assumption of a homogeneous field distribution.

Keeping d/λ constant Paschen's law seems to allow a linear extrapolation from the breakdown strength of gases under normal conditions up to the strength of solid and liquid insulators. If we take the breakdown strength of air at atmospheric pressure, 30 kv/cm, as representative of the gases and the average density of liquids and solids as corresponding to ~ 2000 atmospheres, the condensed insulators should stand about 6.10^7 volt/cm. Actually our normal insulators fail between 10^5 to 2.10^6 volt/cm, leaving us with the task of explaining this discrepancy.

2. The Friction Losses of the Electrons

(a) Excitation of the Electronic System

In air at atmospheric pressure the electrons have a mean free path λ of $\sim 10^{-4}$ cm; hence several volts of kinetic energy can be gained between successive impacts, if the electric field approaches the breakdown strength. Nevertheless the ionization in the field A below the Paschen curve (Fig. 1) remains insufficient because a very effective barrier exists against the acceleration of electrons. Inelastic impacts between electrons and gas particles occur transferring the accumulated kinetic energy of the electrons into excitation energy of the electronic

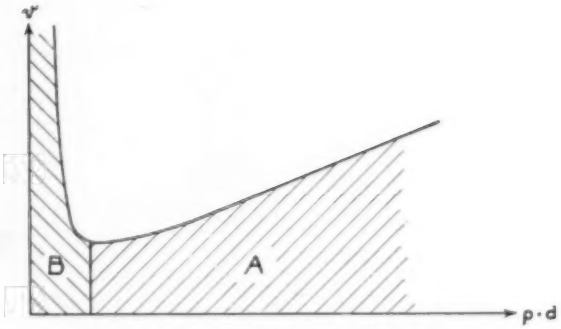


FIG. 1. Paschen's breakdown curve for gases.

system of the particles. This energy, used for radiation, dissociation or in impacts of the second kind, will normally be lost for the ionization process. The probability of such inelastic impacts is a relatively large one, because many transitions below the ionization potential exist. Each one of these quantum transitions will be excited corresponding to its "excitation function" (Fig. 2) which is a plot of the excitation probability against the kinetic energy of the electrons.

This mechanism of friction, protecting the gases very effectively against early breakdown, loses much of its importance in the condensed phases. The influence of the neighboring particles lowers their ionization potential so much that excitation of the electronic system becomes mostly identical with ionization.[2] The classical analogy to this situation is the fact that the energy to dissociate a neutral particle into two charged ones is reduced proportionally to the dielectric constant of the embedding medium.

(*b*) *Excitation of Vibrations*

Below the barrier of excitation of the electronic system just described there lies in molecular gases a second one stopping efficiently slow electrons. Inelastic impacts occur, exciting vibrations. To judge the importance of this type of friction for the condensed phases, we have to discuss the mechanism more in detail.

A slow electron of about one electron volt impinging upon the heavy mass of a molecule would according to the classical impact laws have no chance to transfer oscillation quanta, in a purely mechanical way. But the electrical interaction of the approaching electron with the molecule can change the binding forces between the atoms in the molecule and start oscillations.[3] Fig. 3 gives the "potential curve" of a diatomic molecule, that is the potential energy of the atoms against each other as function of their distance. The minimum of the curve corresponds to the equilibrium position of the nonoscillating molecule.[4] If the approach of the electron distorts this curve, the atoms will tend to move to a new equilibrium distance and may find themselves, after the electron has disappeared, in a new vibration state.

The probability curve for this type of energy

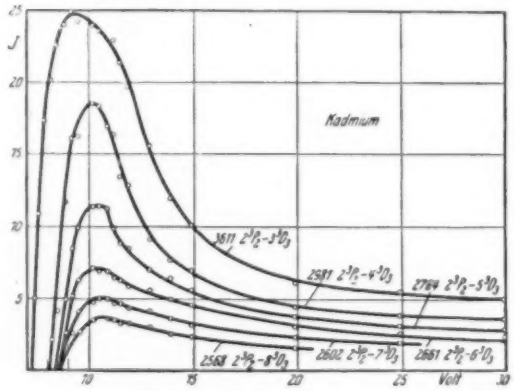


FIG. 2. Excitation function of some cadmium lines. (K. Larché.)

transfer will have a shape as in Fig. 2. It must start with zero, when the kinetic energy of the electron is just sufficient to excite the vibration state in question; it must decrease with increasing velocity of the electron, because the time of interaction becomes small compared with the oscillation time of the nuclei. Hence the "excitation function" steps over a maximum not far above the energy quantum to be transferred (Fig. 4). The yield will be the higher the more the potential curve is distorted; that means the stronger the coupling between electron and molecule. It is therefore bigger for instance for CO, a molecule with a permanent dipole, than for N_2 which has not a dipole moment, though the electronic system in both cases is the same.[5] From the part of the curve measured one can estimate that the maximum yield in CO will be of the order of 20 percent.

This type of friction is not important for the breakdown strength of gases, because the electrons already gain across one free path so much

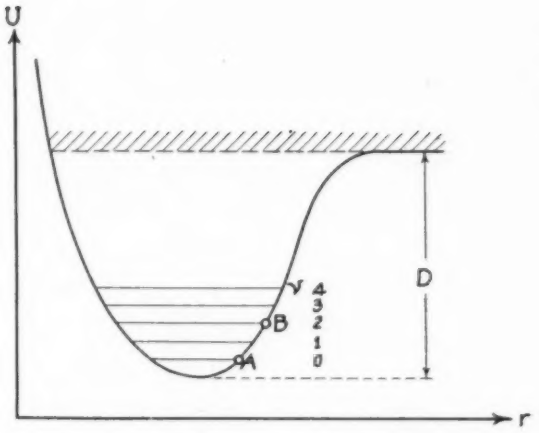


FIG. 3. Potential curve of a diatomic molecule.

energy that they are far above the maximum of the excitation function. But for the condensed systems the situation is quite different. Here the electron is not only coupled with a few atoms as in a molecule, but with a great number of particles, and it is therefore not only able to start oscillations of two particles against each other but to excite more or less the whole spectrum of elastic vibrations of the condensed body. Hence the barrier of oscillations has been broadened very much and has gained in intensity in the condensed phases, while the barrier of excitation of the electronic system has been widely destroyed. We should therefore expect that the friction losses by excitation of vibrations determine now the breakdown strength.[6]

According to Debye's treatment of the specific heat[7] we represent the system of elastic vibrations by a continuous band of frequencies extending from zero to ν_{max} (Fig. 5). The density z of occupation in this band increases with the frequency as

$$dz \sim \nu^2 \cdot d\nu. \qquad (1)$$

Each of these vibration levels has an excitation function like Fig. 4, the whole band therefore an integral excitation function (cf. Fig. 5) representing the sum of the individuals. In analogy to Fig. 4 and J. J. Thomson's treatment of the ionization of atoms by electron impacts[8] we can expect that the maximum of the integral excitation function lies about twice above the minimum energy sufficient to excite the limiting frequency ν_{max}.[9]

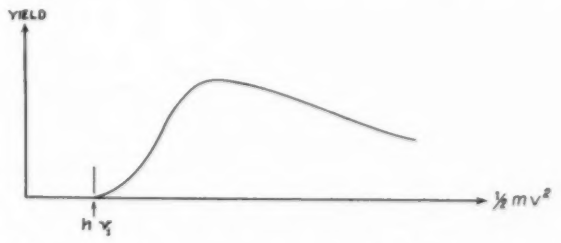


FIG. 4. The excitation function of the vibration state ν_1.

3. The Breakdown Strength of the Alkali-Halide Crystals

The general picture just developed, that the barrier of vibrations is the main protection against breakdown in the condensed phases, can be tested in detail on the single crystals of the alkali-halides. They are very good insulators, crystallize in a very simple ionic bound lattice (Fig. 6), the limiting frequency ν_{max} of their elastic spectrum is known and besides this spectrum of traveling waves there exist only the standing vibrations of the positive partial-lattice against the negative one, the "Reststrahl" frequencies, which practically coincide with ν_{max}. Hence the position of the maximum of the integral excitation function is approximately known (cf. Section 2).

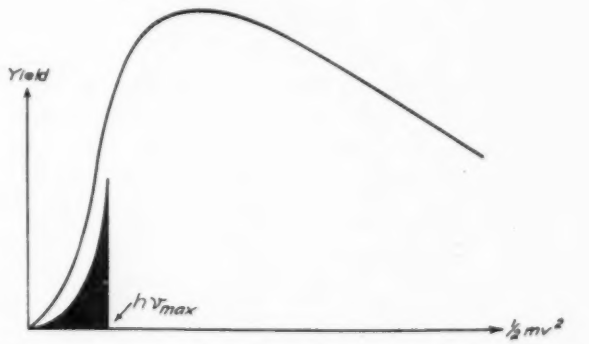


FIG. 5. The spectrum of elastic vibrations and its integral excitation function.

Electrons traveling through these crystals with kinetic energies below the maximum of the excitation function will very likely lose their energy again and again in inelastic impacts, exciting vibrations; falling in an electric field they follow Stoke's law of friction, that is Ohm's law, if sufficient electrons are available. But if they cross the maximum, the friction decreases with increasing velocity, acceleration becomes likely and will not stop until the electrons have accu-

mulated sufficient energy for impact ionization.

Cumulative impact ionization in gases is not dangerous as long as the positive ions traveling backwards to the cathode cannot liberate a sufficient number of electrons to maintain the discharge. This "equilibrium condition," defining breakdown in gases, becomes invalid in the condensed systems; this is a second reason for the failure of Paschen's law (cf. Section 1). In solid insulators for instance the positive ions are more or less fixed in their positions, but the electrons are swept out by the electric field towards the anode, leaving behind the corresponding positive space charge. The space charge spoils the primary field distribution and weakens the structure of the material, in both ways favoring the progress of the ionization process. Hence the start of cumulative impact ionization in the condensed systems is the start of breakdown, if the process is not brought to a stop by special means.

This situation allows one to predict the breakdown strength of the alkali-halide crystals. Breakdown will occur if the electric field applied becomes so high that electrons traveling through the crystal have a sufficient chance to traverse the maximum of the integral excitation function. An electron migrating through the lattice is very strongly coupled with the ions of the crystal. If in CO the maximum yield reached 20 percent (cf. Section 2) we must expect about 100 percent probability as maximum yield in the alkali-halides. The distance between two neighboring ions represents the distance between two successive impacts; across this distance $a_w/2$ (cf. Fig. 6) the electrons must therefore gain sufficient energy to make acceleration likely. If we call $E_{\max}$ the breakdown field strength, then $e \cdot E_{\max} \cdot a_w/2$ is the energy gain in question; it must reach or even surpass the energy $C \cdot h\nu_{\max}$, which corresponds to the maximum of the integral excitation function, if impact ionization and breakdown shall occur.

$$e \cdot E_{\max} \cdot a_w/2 \sim C \cdot h\nu_{\max}. \qquad (2)$$

The factor C must have a value as shown above (cf. Section 2), in the neighborhood of 2.

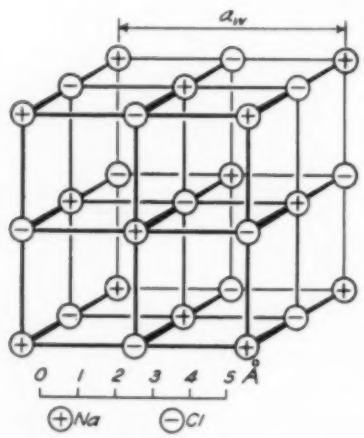


FIG. 6. The lattice of rocksalt.

TABLE I. *The relation between breakdown strength and infrared vibrations for the alkali-halide crystals.*

CRYSTAL	$e \cdot E_{\max} \cdot a_w/2$ (10^{-2} volt)	$h \cdot \nu_{\max}$ (10^{-2} volt)	C
LiF	6.3	7.2	0.9
NaF	5.6	3.3	1.7
NaCl	4.3	2.4	1.8
NaBr	2.5	1.8	1.4
NaI	2.3	1.6	1.4
KF	4.8	3.0	1.6
KCl	3.3	1.9	1.7
KBr	2.3	1.5	1.5
KI	2.1	1.3	1.6
RbCl	2.7	1.7	1.6
RbBr	2.0	1.15	1.7
RbI	1.8	1.05	1.7

Figure 7 shows the breakdown strength $E_{\max}$ of the alkali-halides, carefully measured on single crystals;[10] Table I confronts breakdown strength and infrared vibrations according to Eq. (2). The factor C lies indeed between 1.4 and 2,[11] showing that the breakdown strength in these cases can be calculated by our theory with an accuracy corresponding to that with which the excitation functions can be estimated.

4. Preferential Breakdown in Special Crystallographic Directions

Our theory of the electronic breakdown can be tested in a second and quite independent way on the crystalline insulators. A surplus electron moving along through the space grating of the alkali-halides (Fig. 6) finds the possibility of becoming accelerated not only dependent upon the field strength applied but also on the lattice direction. The three main directions of the rocksalt structure, seen from the standpoint of the electron (Fig. 8) differ in two aspects. The distance between the ions is different and the hills

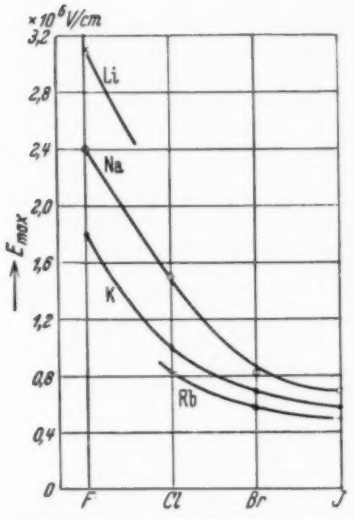


FIG. 7. The breakdown strength of the alkali-halide crystals. (The last letter on ordinate should be I instead of J.)

and valleys of the potential structure differ in shape and elevation. While of course the absolute values of the altitude are not known, their qualitative picture can be taken from the model. It shows the path with the lowest potential hills stretching in the direction of the face diagonal [110], the path with the smallest number of ions pointing towards the room diagonal [111], while the cube edge [100] is marked by densest occupation and steepest potential variation.

According to our assumptions (cf. Section 3) breakdown occurs if electrons in the lattice become sufficiently accelerated between two impacts to cross the maximum of the excitation function and then gain further energy up to about 10 ev, until they can knock out the electron of the negative halide ion. Repeating the process again and again the electrons form electronic avalanches sweeping through the crystal towards the anode and leaving behind a track of neutral halide atoms and positive alkali ions, a positive space charge. Now it can be seen from the slow decay of the integral excitation function (Fig. 5), that even behind the maximum the friction is very great and that therefore the acceleration of the electron must be first very slow. Consequently the path on which the electron will gain its first 1 ev of kinetic energy will be a long one compared with the distance along which it gains the rest of the energy needed for impact ionization. The slow moving electrons determine the general direction of the path which the breakdown process follows.

Figure 8 shows that the face diagonal [110] offers the lowest potential hills to climb; this direction is therefore the first one to be passable for slow electrons and the electric breakdown at the minimum potential should be expected to take this path. If, however, high overvoltages are applied the bigger hills of the room diagonal [111] can now be overcome after a short distance of acceleration. Since this [111] direction offers the smallest number of impacts and hence the smallest friction it will now be chosen by the breakdown process.

This crystallographic orientation of the breakdown path in a "first" and a "second preferential direction" can be made experimentally visible, because the lattice collapses along the space charge track.[12] By using point electrodes and a heated crystal the whole breakdown process can be directly followed up in its development.[13] From the anode a path breaks forth in [110] direction and proceeds, bending and branching in face diagonals on its way towards the cathode (Fig. 9). The corners of the track, first definitely rectangular, become rounded under the heating influence of the current, small side branches melt away again and a path of destruction remains, reminding one of its origin by the sinusoidal track (Fig. 10), or even covering its genesis perfectly under the smoothing influence of the current (Fig. 11).

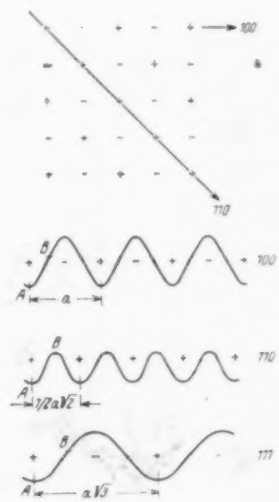


FIG. 8. The potential structure of the rocksalt lattice from the standpoint of a surplus electron.

The experiment just described reveals only the "first preferential direction of breakdown," because the geometrical arrangement of the electrodes avoids higher overvoltages. Only in the neighborhood of the point electrodes is the crystal subjected to breakdown stress (Fig. 12). The tracks of positive space charge, formed by successive electronic avalanches, act as assistant electrodes and bridge the gap step by step. If we start with an homogeneous field distribution breaking down thin plates of single crystals between plate electrodes, the situation is very different. Here we put the whole volume under breakdown stress from the beginning. The development of a space charge track changes the geometry of the field rapidly to a point against plate arrangement (cf. Fig. 12) resulting in high overvoltages. Hence the breakdown develops very fast (in about 10^{-7} sec.)[14] and its steps can only be reconstructed afterwards by a microscopical examination of the sample if sufficient precautions are taken against secondary destruction.[15]

This second experiment now confirms the existence of a "secondary preferential direction of breakdown" as expected above. From the anode plane of the crystal the track starts as before, diving into the plate in [110] direction toward the cathode (Fig. 13). But at some depth the path suddenly jumps into the room diagonal direction and follows this way down to the cathode plane. The track is likely to branch into several of the four [111] directions offered and between them the crystal tends to crack in the cleavage direction [100], thus forming very characteristic four-cornered pyramids with the cathode plane as base and the room diagonals as edges (Fig. 14).

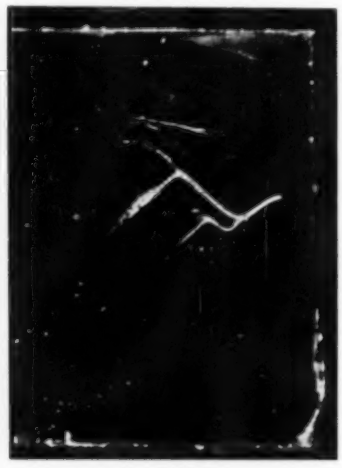


FIG. 9. The breakdown path in development.

The direction laws of breakdown just stated will not only depend on the structure type of the crystal in question but also on the accuracy with which the crystal corresponds to the point model of its atomphysical composition. In the alkali-halide structure for instance (cf. Figs. 6 and 8) the size of the ions and their polarizability increases from Li^+ to Cs^+ and from F^- to I^-. Hence we can expect that the rigid structure formed by the small ions shows especially well the attribute of our model, that is, the pronounced influence of the crystallographic direction. In the soft crystals of the big ions the charge distribution becomes more and more blurred, therefore the direction of the applied field will be of higher importance.

TABLE II. *Appearance and direction of the breakdown path in the alkali-halide crystals.*

CRYSTAL	APPEARANCE	DIRECTION (the field direction is [100])
LiF	sharp	primary [110]
NaF	"	secondary [111]
NaCl	"	
KCl	"	[110] and [100]
RbCl	"	
NaBr	a little blurred	[110], but also [111], [100] and other directions
KBr	blurred	[100] and sometimes branching
KI	"	
RbBr	"	
RbI	"	

Table II summarizes the results of breakdown tests obtained on plates of single crystals in homogeneous field arrangement; they confirm the influence of the ion type on the direction of breakdown. Hence our theory of the electronic breakdown seems well supported. In its special application to crystals, the one integral excitation function of Fig. 5 has to be replaced by a number of such curves, each one characterizing the friction in a special crystallographic direction. Fig. 15 illustrates schematically the situation for rocksalt according to the experiments described.[16]

FIG. 10. Sinusoidal breakdown track in KBr.

5. *The Capture of Electrons*

Our discussion with all its consequences is based until now on the assumption that there are surplus electrons in the insulator which can be accelerated. Now the existence and the origin of such electrons has to be established.

If we start with an electron created by some photoelectric action and diffusing through the insulator, we could expect (cf. Section 2), that it would soon float in thermal equilibrium with its surroundings, slowing down by transferring and speeding up by absorbing vibration quanta statistically. But this viewpoint would only take into consideration the kinetic energy of the electron, whereas the decisive factor in the excitation process is the coupling between the electron and its surroundings, that is, the potential energy. Hence the electron can lose more than just velocity in migrating along; it can also lose potential energy and become trapped.

The general condition for a capture process is that by the recombination process energy becomes liberated and dissipated before the particles have time to separate again. Hence if no dissociation or radiation provides an outlet for the energy the capture of an electron is only possible in a "three body impact." A third body participates in the collision acting as energy acceptor. This condition makes recombination in gases generally improbable but is automatically fulfilled in the condensed phases on account of the coupling between the particles. Therefore every electron which is traveling slow enough to permit the transfer of its surplus energy into vibration energy of the heavy particles becomes attached.

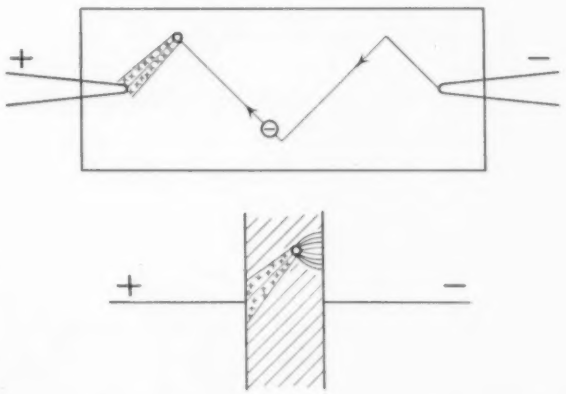


Fig. 12. The development of breakdown in inhomogeneous or homogeneous field arrangement.

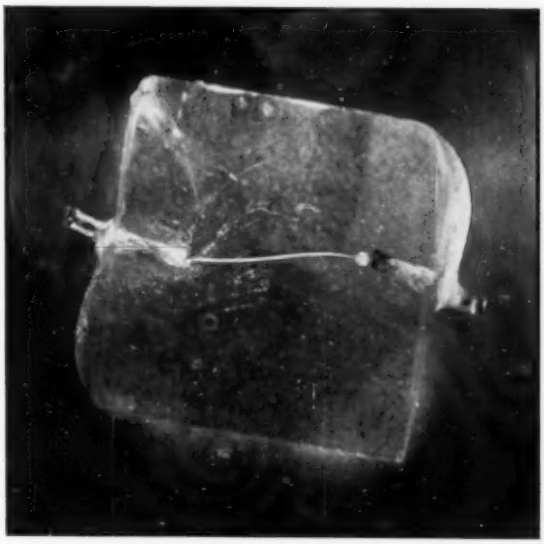


FIG. 11. Smooth breakdown track in KBr.

In discussing the potential curve of a diatomic molecule (Fig. 3) it was stated that oscillations can be excited, if the heavy nuclei have sufficient time to move during the distorting action of the electron. Taking the same standpoint for the condensed systems we can formulate the condition of capture. The shortest oscillation time of the masses, if intramolecular vibrations do not play a role, is $t=1/\nu_{\max}$; the time for the electron to travel from one particle to the other across the distance l is $T=l/v$. Capture becomes likely if T reaches or surpasses the fraction t/A, where the

factor A depends on the intensity of the coupling between the electron and the particles. Calling this critical velocity of the electron v_k we may write:

$$v_k \leqq A \cdot l \cdot \nu_{\max} \tag{3}$$

or in special application on the alkali-halide crystals, where

$$l = a_w/2, \quad v_k \leqq A a_w / 2\nu_{\max}. \tag{3a}$$

A must be larger in an ionic structure like rocksalt than in an atomic one like diamond, or in a molecular liquid like insulating oil. Assuming A as about 60 for NaCl, where $\tau = 1.7 \cdot 10^{-13}$ sec. and $a_w = 5.6 \times 10^{-8}$ we would expect that in this crystal electrons already having a velocity of 0.02 ev become captured.

Every electron bond produces an optical absorption corresponding to the photon energy needed for its dissolution. Hence the behavior of slow electrons in insulators can be studied not only by electrical measurements but also by the absorption spectra, which correspond to the capture processes. For the alkali-halide crystals a very large experimental material is already available, which we owe especially to R. W. Pohl and his co-workers at Göttingen. The following discussion uses these observations to control and complete the picture of the capture process just drawn.

6. *The Migration of Surplus Electrons Through the Crystal-Lattice of the Alkali-Halides*

Pohl and co-workers, liberating electrons in the alkali-halide crystals by irradiation with ultraviolet light, found that as the result a new absorption band appeared in the visible spectral range (Fig. 16), the "color" or "F" band.[17] The "color centers" formed in this way or by other means are ascribed to alkali atoms; the fact that light absorption in the F bands originates a photoelectric current supports this viewpoint. But Pohl and other authors[18] assume that the color centers form by recombination between electrons and positive ions on special deranged points of the lattice on "Störstellen," [19] while our theory explains the capture as a consequence of the structure of the normal crystal. The following discussion will show that the observations of Pohl and his co-workers seem to be in favor of our point of view.

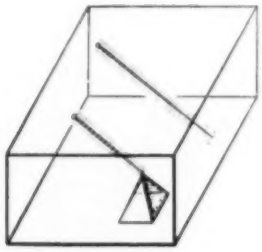


FIG. 13. Scheme of breakdown through a rocksalt plate.

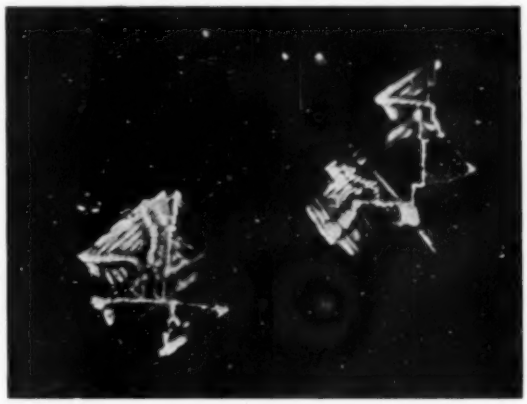


FIG. 14. Microscopic picture of the pyramid structure of the breakdown through a rocksalt plate.

A surplus electron put in the ideal lattice of rocksalt finds according to Fig. 8 the different directions in the crystal differently favorable for its migration. The space grating is composed of potential cups surrounding the positive ions and separated from each other by walls of different height and steepness; the structure of the lattice plane [110] illustrates the situation. (Fig. 17.) The electron moving from one cup to the other in statistical motion will normally escape over the lowest points of the separating walls, that is, in one of the 12 face diagonal directions. If the structure would be rigid, the potential humps extending in these directions could not form a serious obstacle at all, as Fig. 18 demonstrates. The electron staying in one potential cup changes the corresponding positive ion in an alkali atom, about doubling the diameter of this particle. This big increase in size has the effect that the atomic charge distribution of the positive ions in the [110] direction practically touches each other.[20] The electron therefore could not belong to a

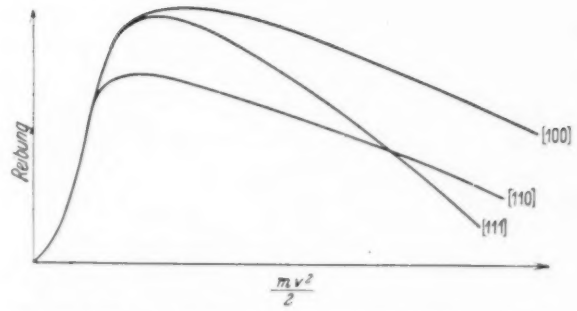


FIG. 15. Scheme of the three integral excitation functions of rocksalt.

single ion but would travel unlocalized in a conducting band stretching itself through the lattice in the face diagonal directions.

Now the lattice is not rigid but starts to dissipate energy of the electron into vibration energy of the crystal (cf. Section 2). If the electron remains so long in a single potential cup that the surrounding ions move toward new equilibrium positions, the cup becomes deformed and widely deepened. It becomes a trap (cf. Section 5). The electron, caught by the distortion of the lattice which it itself has brought about, is perfectly localized.

The type of binding which keeps the surplus electron in place is according to Fig. 18, in the alkali-halide crystals, a very unusual one. The electron, in recombining with a positive ion, forms an atom of such a large size, that the twelve positive neighbors participate on the charge with practically equal rights. Hence the electron is really not bound by the central particle but by a group, which again is a part of the lattice. Every displacement, such as heat vibration, especially if it affects one of the 18 next neighbors, influences directly the binding energy of the electron.

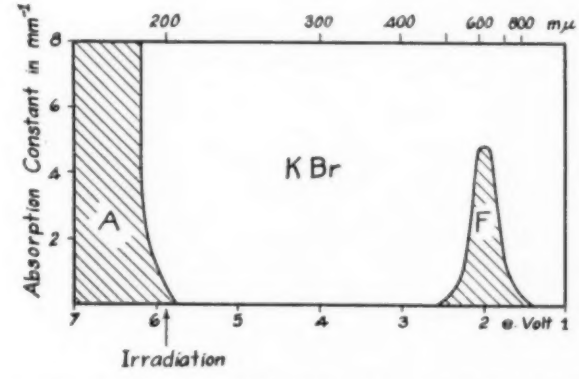


FIG. 16. Formation of the F Band by irradiation in the ultraviolet absorption A.

This conception of the nature of the atomic color centers demands that the color band representing the binding energy of the surplus electron should be very sensitive against changes of the crystal temperature.[21] That is the case as shown by the measurements of Mollwo[22] (Fig. 19) or by the direct observation that a rocksalt crystal with color centers turns from pink to yellow in cooling down from 500°C to room temperature. The shape of the color band reacts equally sensitively with mechanical stresses as the work of R. Przibram and A. Smekal indicates.[23]

The even more important consequence of this

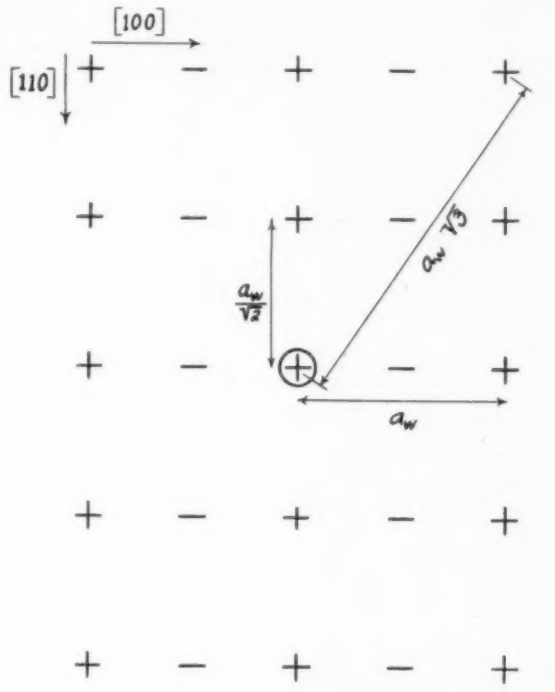


FIG. 17. Distances from one potential cup to the next one in the [110] plane.

type of "group-binding" is that a concerted action of a number of ions each having only an average heat energy of about 1/10 ev at its disposal, may deliver volts of activation energy for the liberation of the surplus electron by adiabatic redistortion of the crystal lattice. This effect makes the thermal mobility of the color centers possible, which is so impressively observed in the experiment first done by Stasiw.[24] The cathode of a sufficiently heated alkali-halide crystal emits

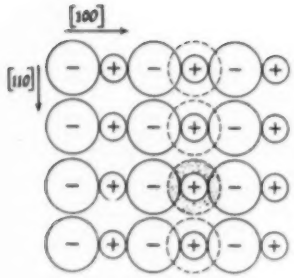


FIG. 18. The ionic radii in the rocksalt crystal and the atomic radius of the Na atom (broken-line circles) in the same lattice.

clouds of color centers traveling to the anode (Fig. 20). Stasiw found, as will be explained below, that the velocity of the centers was proportional to the field strength applied and depended on the temperature in a peculiar way (Fig. 21). Using high temperatures a defined velocity curve can be measured, but in the lower temperature range the curve broadens into a field of values containing very high ones too.

Pohl and co-workers studying the mobility of the electrons used according to their conception of the color centers the following picture: The electrons move through the crystal until they stick on a "Störstelle." After some time they are thermally liberated and so on. The "mean displacement w" in field direction between two successive capture acts has been measured first by Flechsig[25] and Hecht.[26] They liberated electrons in a small region of a cooled crystal by a narrow light beam and measured electrically the decay of the number of electrons as function of the distance x from their origin and of the field strength applied. The authors found the statistical law

$$N = N_0 e^{-\alpha x}, \tag{4}$$

where the absorption coefficient $\alpha = 1/w$. The

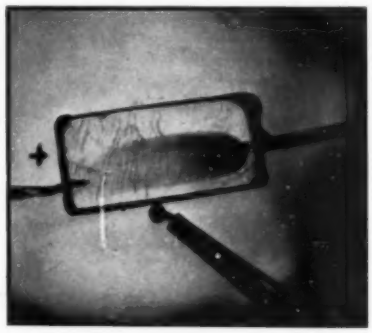


FIG. 20. Color centers in rocksalt traveling to the anode.

"mean displacement w" increased proportionately to the field strength. Smakula[27] later measuring the velocity c of the color centers in a heated crystal by optical and electrical means, found that the time of capture is determined by a Boltzmann law:

$$c = c_0 \cdot e^{-U/kT}. \tag{5}$$

The whole situation just described is easily understood by our theory of the binding mechanism. The average velocity c of a color center in field direction consists of two parts: the electron travels freely during a time t_f across the mean capture distance w and stays a time t_b bounded in the lattice

$$c = \frac{w}{t_f + t_b}. \tag{6}$$

The time t_b of capture will be infinite if the temperature is zero, and will correspond to the shortest oscillation time τ (cf. Section 5), if a sufficiently high temperature immediately places the activation energy U in form of vibration energy of the lattice to the electron's disposal.

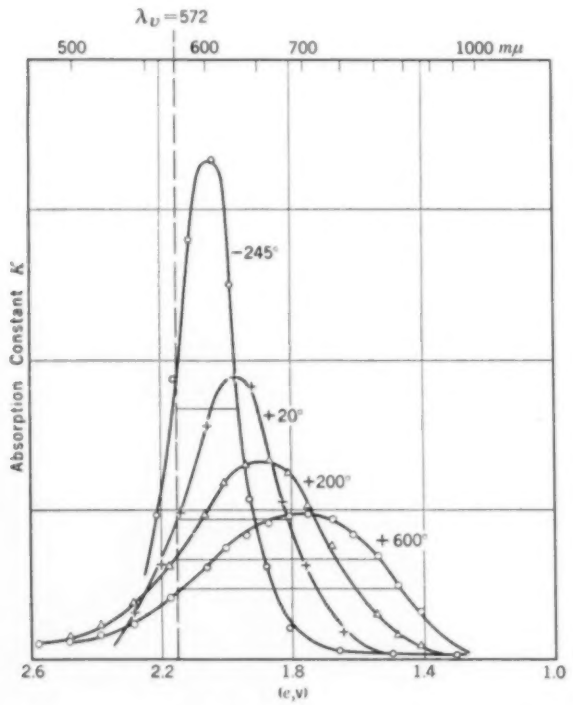


FIG. 19. The temperature sensitivity of the F bond in KBr. (Möllwo.)

Between both these extremes a Boltzmann distribution law exists

$$t_b = \tau \cdot e^{U/kT}. \tag{7}$$

Now, if w is not large, and this is normally the case in the alkali-halides, $t_f \ll t_b$ and (6) takes the form

$$c = w \cdot \nu_{\max} \cdot e^{-U/kT}. \tag{8}$$

The velocity c of the color centers is found proportional to the field strength since the average velocity of the electrons increases with the field strength and therefore according to (3) the capture becomes delayed and w increases. It is hard to see how a "Störstellen" mechanism could without artificial assumptions explain the influence of the field strength.[36]

We should expect (cf. later Section 8) that w increases with decreasing temperature. This, and the fact that the liberation act takes more time, could account for the lower end of Stasiw's curve (Fig. 21). The statistical character of w blurs the boundary of the cloud and gives a velocity distribution. The connection between c and $\nu_{\max}$ cannot directly be observed, because U will depend on $\nu_{\max}$ too.

7. *Short Review*

It seems advisable now to stop for a moment and to review what we have accomplished and what remains to be done.

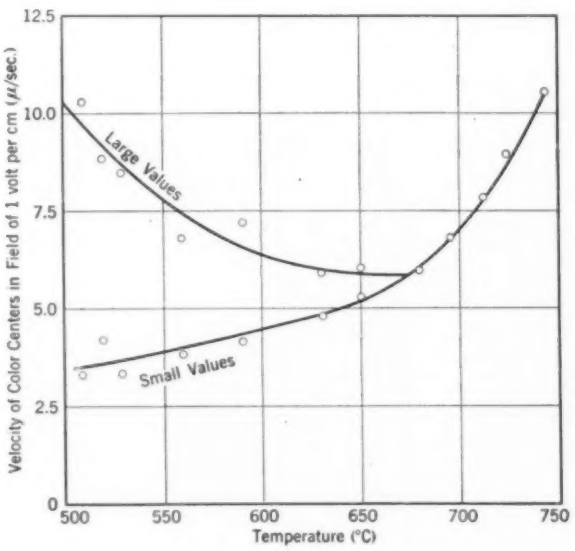


FIG. 21. Velocity of the color centers in KCl. (E. Stasiw.)

We have found four reasons why the simple extrapolation of Paschen's law in gases towards the breakdown strength of solid and liquid insulators must fail.

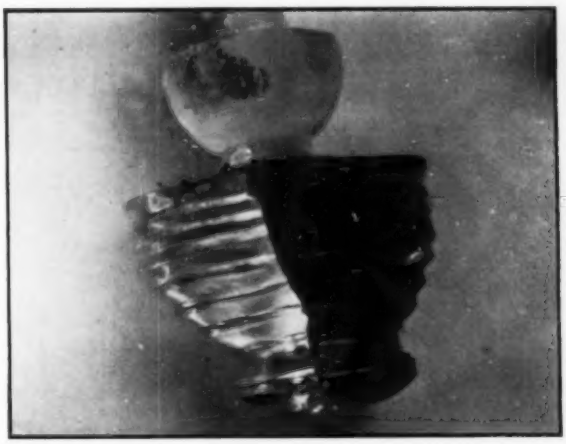


FIG. 22. Two crystals of KBr grown simultaneously, the right one colored blue by electrons.

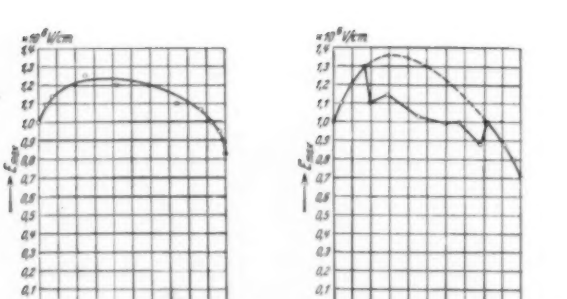
1. The friction losses of the electrons preventing early cumulative ionization, have changed; instead of the excitation of quantum transitions in the electronic system of the particles the excitation of vibrations becomes decisive (cf. Section 2).

2. The breakdown condition in gases stating that the discharge started by electrons must regenerate sufficient electrons for its maintenance (equilibrium condition) becomes invalid in the condensed phases (cf. Section 3). Electrons can be supplied by electronic emission of the cathode (cf. Section 6).

3. Cumulative ionization creates positive space charges which play a dominating role for the development of the breakdown by distorting the field distribution and weakening the material (cf. Section 3–4).

4. Slow electrons in the condensed phases normally become captured (cf. Section 5–6).

We have shown that the breakdown strength can be predicted and have proved it for the alkali-halide crystals (cf. Section 3). Structure effects offered by the simple composition of this material allowed us to study the breakdown process in every detail and to confirm the theoretical conclusions (cf. Section 4). A special study of the behavior of slow electrons stressed the importance



(a) (b)

FIG. 23. Breakdown strength as function of the composition.
(a) KCl—RbCl; (b) KCl—KBr.

of the capture process in condensed systems (cf. Section 5), revealed that for the alkali-halides the atomic "color centers" correspond to surplus electrons held by a new type of "group binding" and completed our picture about the migration of electrons in these crystals (cf. Section 6).

But now a last step has to be taken: The use of the alkali-halides as test material can be objected to because the characteristic feature of a perfect periodic structure is normally missing in technical insulators. Hence we have to eliminate now the influence of this factor on the breakdown process by spoiling systematically the order in our crystals, by progressing from the crystalline to the glass state. Then we will get the general picture of the electric breakdown in solid and liquid insulators.

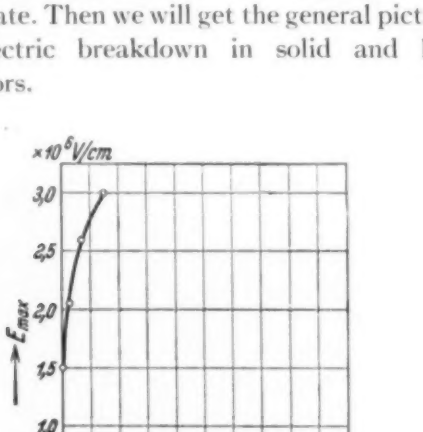


FIG. 24. Increase in the breakdown strength of rocksalt by admixture of silver ions.

8. *Disturbing of the Crystal Lattice and the Consequences for the Capture Process*

Starting towards this goal we disturb the crystal structure of the alkali-halides in two definite ways and observe the experimental result:

a. We spoil the periodicity abruptly by inserting an interior boundary. This can be done most

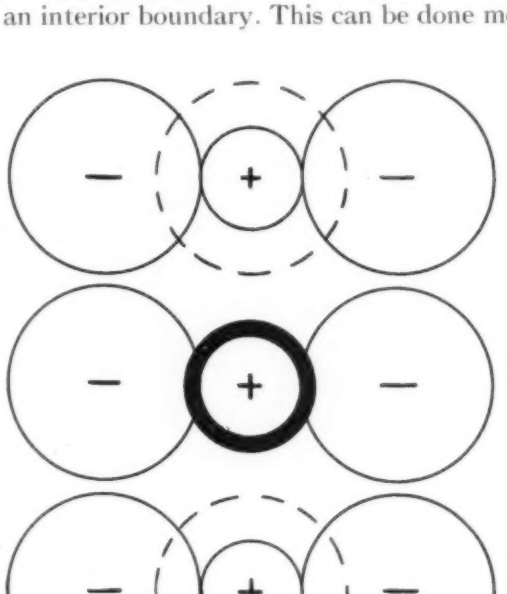


FIG. 25. Copper atom (center) and sodium atom (above and below) in the rocksalt lattice.

naturally by growing two single crystals simultaneously in the same block of material.[28] During this process, while the crystals are in intimate contact and heated just below the melting point, electrons are released from a point cathode into one or the other of the partners. As Fig. 22 demonstrates they cannot penetrate through the boundary; the KBr crystal, which is directly infected, turns blue, the other one stays clear without color centers.

b. We disturb the periodicity more progressively by growing single crystals, which contain increasing admixtures of foreign ions. Studying the influence on the breakdown strength of the material we get curves[29] which pass over a maximum like the resistivity characteristics of metal alloys (Fig. 23). The irregularities of the KCl–KBr curve may indicate as in alloys the

formation of a "superstructure." A most spectacular increase of breakdown strength is achieved in rocksalt by the admixture of silver ions (Fig. 24).

To discuss these facts we resume some considerations of Section 6. A surplus electron in the ideal crystal lattice could easily escape from one potential cup to the other across one of the twelve low potential walls in the [110] direction. Every disturbance of the structure will raise some of these barriers and may lower others. This will cut down the stereo-probability of escape for the electrons which could pass before. Hence the probability for capture increases and the "mean displacement w" shortens. Measurements of Hecht on AgCl which has the same structure show directly that w is dimidiated by addition of only 0.05 mole percent CuCl.[30] Thermal disorder acts in the same direction as the dependence of the velocity of the color centers on the temperature (Fig. 21) seems to indicate.

Increased probability of capture is not the only result of the lattice distortion but the binding energy also becomes affected (cf. Fig. 19). The whole type of "group binding," based on the large size of the alkali atom and the competition of the neighboring ions (cf. Section 6) may be destroyed and the behavior of the attached electron completely altered, as the following example shows. Some of the Na^+ ions of a rocksalt crystal shall be replaced by Cu^+ ions; this exchange does not alter the situation so far as the ionic radii are concerned (Cu^+ 0.95 A.E.; Na^+ 0.96 A.E.). But for the fate of a surplus electron traveling along the aspect is altogether changed. While the sodium ion in attaching the electron would take up an atomic radius of 1.85 A.E., the copper ion is binding the electron on account of the high ionization potential in a very narrow radius of 1.22 A.E. Hence copper ion and electron have practically recombined (Fig. 25); the surplus electron has been withdrawn so far from the neighbors that their influence is only a disturbance of the second order. The copper atom becomes a foreign substance in the crystal forming an interior boundary in the lattice and connected to it by adsorption forces.

This breaking of the link between lattice and surplus electron has the decisive consequence that the new color center formed has lost its thermal mobility. The electron does not profit any more directly by the heat vibrations of a group, because the main part of its binding energy comes from a single ion and sufficient heat energy for liberation had to be stored at this single point. This process would be so unlikely for the temperatures in question that we have to expect a color band (cf. Section 6) insensitive to temperature change and a color center fixed in position.

Our conclusions can be verified experimentally for the case just treated: We seal two copper wires in a rocksalt crystal, place the sample in an electric furnace (Fig. 26), heat up to about 500°C

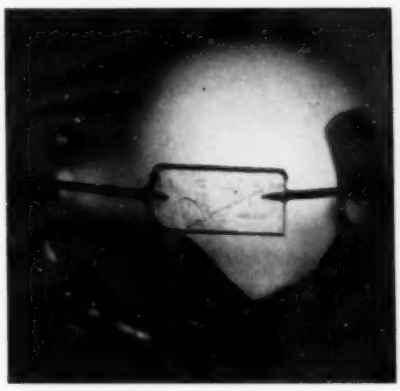


FIG. 26. The sample in the electric furnace.

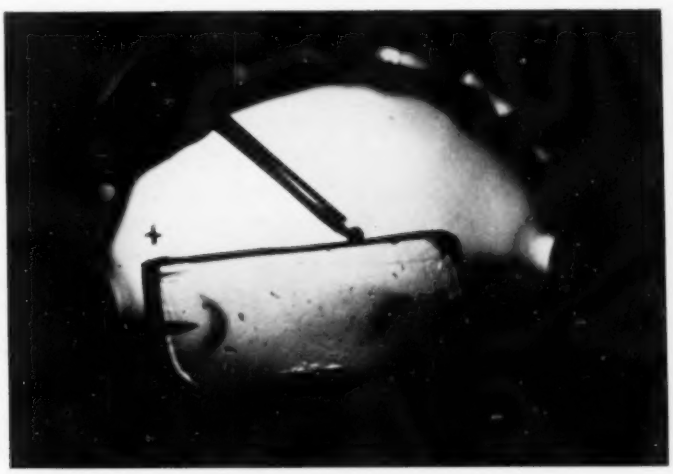


FIG. 27. Developing of the copper ions by electrons in the anodic part of a rocksalt crystal.

FIG. 28. Dendrite tree in [110] direction. (NaCl with 1 percent AgCl, below 570°C.)

and put a small voltage between the electrodes to electrolyze some copper ions into the crystal. After some time we raise the voltage and sweep electrons from the cathode as color centers through the rocksalt (cf. Fig. 20). Approaching the anodic part of the sample the electrons suddenly develop a ruby color in the crystal which hinders the further progress of the electrons appreciably (Fig. 27). This color does not move in the electric field. It does not change visibly with the temperature of the crystal and is even about the same, if any other of the alkali-halide crystals is used for this experiment.[13] The red boundary lies more toward the cathode if we start the electrons later. The color centers are copper, as the chemical analysis confirms.[31] The electrons have acted like a photographic developer and recombined with the copper ions in the expected manner.

The case of the sodium atom and the copper atom in the alkali-halide lattice represents the two extremes of "group binding" and "atomic binding." Between them all intermediate states are possible. A sodium atom on a crystal surface for instance is only coupled with about half of the original group and will therefore have a longer lifetime in the bounded state. A colloidal complex of atoms is already a foreign substance in the lattice. Pohl and his co-workers observe that the corresponding bands are independent of the temperature, as we would expect.[32]

Now we can understand the experiments which we described in the beginning of this section. The electrons did not penetrate through the crystal boundary (cf. Fig. 22) because they are captured like surface electrons. If they are liberated by the heat vibration of the lattice they can only go back in the interior of that crystal to which they belong. The strong action of the silver ions on the other hand is due not alone to the capture process itself which corresponds to that of the copper ions, but to the creation of interior boundaries by the recombination process which stops succeeding electrons.

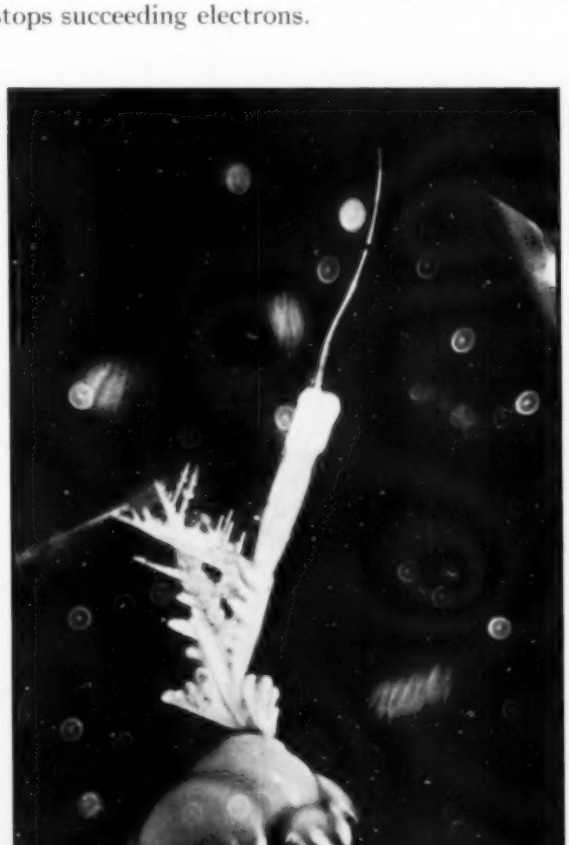


FIG. 29. Dendrite developing in [110] direction. (NaCl with 1 percent AgCl, above 570°C.)

9. *From Crystal to Glass*

This knowledge gained allows us to prepare test material in which one special characteristic of its crystalline state is eliminated. By adding 0.1 percent AgCl to NaCl the mobility of the surplus electrons slows down very much. 1 percent AgCl suppresses the migration of the color centers perfectly—the influence of the "group binding" is destroyed!

We take this material, seal Pt wire in as electrodes and observe in the mounting of Fig. 26 what happens if we increase the field strength. The crystal first stays clear, but as we approach breakdown strength in the neighborhood of the point electrodes, two things happen. From the anode path there breaks out again a path in the [110] direction of the cathode (cf. Fig. 9). Hence the mechanism of electronic breakdown has not changed. Out of the cathode and almost as fast, grows a tree, a dendrite, toward the anode. This tree grows and branches below a temperature of about 570°C in the [100] direction (Fig. 28); above this temperature in the [110] direction (Fig. 29).

This dendrite phenomenon was already found by the author in clean rocksalt[13] and ascribed to the discharging of cations by electrons. The new observations in NaCl–AgCl crystals allow a finer analysis of the situation. The trees in these crystals show the reddish-brown color which is characteristic for silver centers developed by electrons (cf. Section 8). A microscopic examination reveals that in the immediate vicinity of the [100] dendrites the crystal has been split up in the cleavage planes [100]. The cracks are colored brownish by electrons which recombined on the surface and accompany the whole dendrite like a shadow (Fig. 30). If we take these results together with the breakdown mechanism as understood in Section 4, the explanation offers itself.

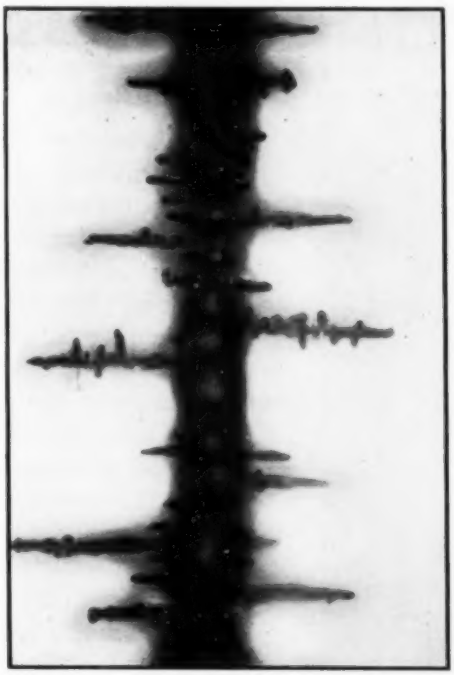


FIG. 30. Part of a [100] dendrite with cracks accompanying the structure like a shadow.

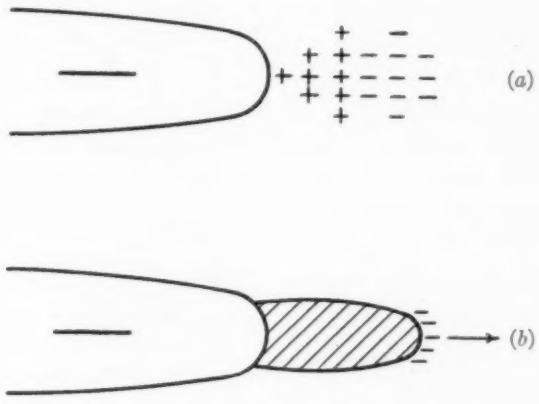


FIG. 31. Breakdown from the cathode side. (*a*) Space charge formation. (*b*) Metallic elongation of the electrode by neutralization and coagulation of the atoms.

When in the neighborhood of the cathode the field strength reaches the breakdown point, an electronic avalanche develops into the crystal leaving a positive space charge behind. This weakens the field strength in front of the electron cloud. The electrons slow down and are trapped by silver ions (Fig. 31). Directly in front of the point cathode the field strength has risen stimulating electron emission until the space charge is neutralized. Again silver atoms and sodium atoms are formed by this process. The atoms coagulate originating a metallic elongation of the cathode. If the field strength is high enough, that means the new point electrode is not less favorable than the original one, the and process starts again. The space demand of the newly formed atoms sets up mechanical stresses in the material. Below 570°C

the crystal gives in by splitting in its cleavage planes, hence the [100] dendrites develop. Above this temperature the crystal is plastic enough to avoid cracking. Therefore appears now in the dendrites too the direction [110], the "first preferential direction" of breakdown (cf. Section 4).

Now we take the final step and destroy the influence of preferential directions by choosing glass as test material. Using first Pyrex glass with Pt point electrodes in the same arrangement as before, heating to about 400°C, we observe: The glass remains first perfectly clear but suddenly, if the voltage is high enough, a brownish cloud shoots up from one point of the cathode wire—and stops. A microscopical investigation shows that a crack has started at this point and its surface has been colored by recombining electrons as shown in Fig. 30. A further increase of the voltage may enlarge the crack in successive leaps. Fig. 23 shows three of them around the electrode. Cracks may also start from other points of the cathode and in reversing the field direction we can start the same phenomenon again from the new cathode. Fig. 33 gives one example. In Pyrex glass the crack stays local. In soda glass the whole sample breaks in pieces.

On the anode side of the glass samples something very different happens. Little sparks begin to appear on the anode wire, hopping irregularly along the surface, disintegrating the glass and growing in size, if the voltage is increased. At last a single spark tends to develop into an arc and to destroy the material.

Both phenomena just described have the same reason: impact ionization by electrons. But while the breakdown could proceed in the alkali-halide crystals because the point structure of the electrodes was preserved, thanks to the existence of a preferential direction (cf. Figs. 9, 28, 29), the starting breakdown in glass is undirected. It alters its course at random, spoils the point shape of the electrodes, lowers in this manner the field strength, and dies out. The cracks on the cathode form because the electrons penetrate into the material where they recombine with ions and set up mechanical stresses. The sparks on the anode side indicate electron avalanches, which leave the crystal while the positive space charge remains behind.

If a homogeneous field is used, the field distribution cannot degenerate as above and the electronic breakdown proceeds as we should expect (cf. Section 4). A breakdown track penetrates the glass plate coming from the anode and often branching towards the cathode (Fig. 34).

FIG. 32. Three successive cracks around the cathode in Pyrex glass.

10. Some General Conclusions

The series of experiments and considerations given seem to establish that cumulative impact ionization by electrons is the principal breakdown phenomenon in condensed systems. In contradistinction to the breakdown in gases the type of friction slowing down the electrons has changed and the equilibrium condition becomes invalid; the distortion of the field and the destruction of the material following ionization is reason enough for progressive breakdown of the insulation. Hence we cannot expect as

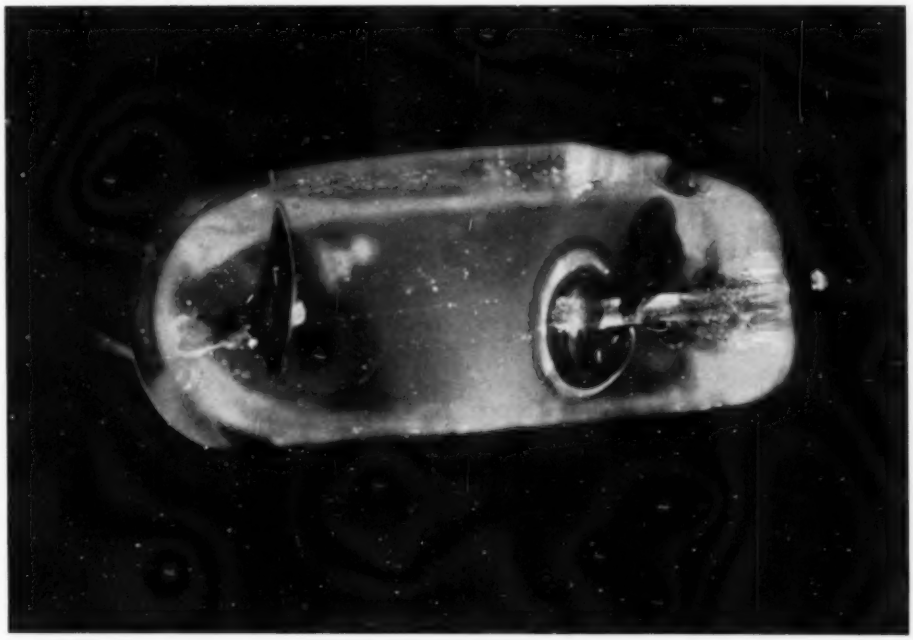


FIG. 33. Pyrex sample with cracks started by recombination of electrons and ions.

Joffe once believed,[33] that the electric strength of insulating materials will increase much with decreasing thickness. There will not exist a field B for the breakdown mechanism as in gases (cf. Fig. 1), in which the strength increases, because the number of ionizing impacts declines below the critical limit drawn by the self-preservation of the discharge. As long as ionization takes place destruction takes place spoiling the insulating properties of the material; hence the intensity of ionization does not mainly influence the breakdown strength but the time to accomplish the final destruction.

The electronic breakdown phenomenon could be followed up to 800°C in rocksalt and about 700°C in glass; surface conduction and the weakening of the material were the only experimental limitation. There is not a new phenomenon "heat breakdown," as it was believed until now,[34] but in every case we have to investigate why the breakdown strength is lowered with increasing temperature. The reasons can be very different as the material here presented already indicates: The electronic emission of the cathode may suddenly start and crack the material on account of recombination between electrons and ions, starting or increasing field emission may distort the field distribution, the increasing mobility of the ions may do the same, dendrites may grow and bridge the gap; in many materials we can expect chemical decomposition, oxidation and so on. "Heat breakdown" is only a word, but not a mechanism; it covers very different phenomena, which we have to know in every case, if we will protect the material against destruction.

Our picture of the breakdown mechanism is developed for the condensed phases in general, it covers therefore the liquid insulators too, but the high mobility of the particles will certainly bring some new features in the situation. If suddenly space charges form by electron emission or impact ionization in the neighborhood of the electrodes, the electrostatic repulsion may overcome the surface tension and bubbles may form in this manner without the presence of occluded gases. Circulation of the insulating liquid on the

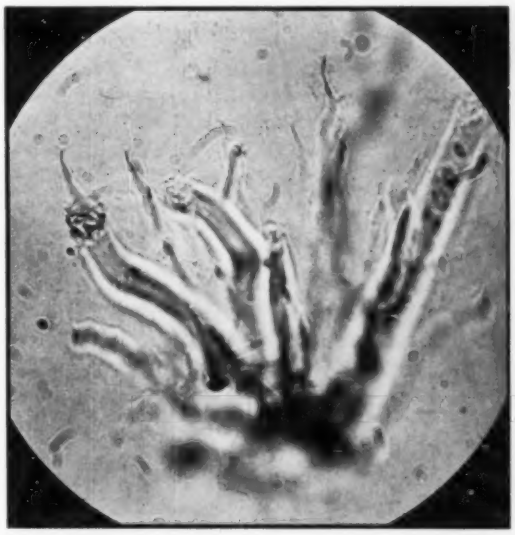


FIG. 34. Breakdown in glass in a homogeneous field. (The picture shows the tracks as shadows coming up to the cathode.)

other hand may destroy field inhomogeneities, which build up slowly and would prove fatal in solids. Some interesting evidence of electronic emission has just been given recently by Baker and Boltz;[35] it is not likely, that the electrons remain free in the cases observed (cf. Section 5). Quite generally a discussion of insulator problems must include the two aspects: Electrons can be charges moving in the electric field but they can also be a most powerful chemical.

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Here and There

Nobel Prize Award

Word arrived November 12th of the award to Dr. Clinton J. Davisson and Professor George P. Thomson, jointly of the 1937 Nobel Prize for Physics. Dr. Davisson, a fellow of the American Physical Society and a member of the Optical Society of America, is a research physicist in the Bell Telephone Laboratories, Inc. The award is in recognition of his pioneering experiments on the scattering of electrons from metal surfaces and the discovery in the course of these experiments that a beam of electrons undergoes diffraction in the same manner as a beam of x-rays. The modern dual view of energy as possessing both a particle and a wave aspect was first established by this result. The technique, aside from its primary importance in enabling the science of physics to take a forward stride, has proved of value in the study of surfaces. Applications have been made to metallurgy and it is anticipated that certain organic films of biological interest can be investigated similarly. Professor Thomson, of London, is the son of Sir J. J. Thomson. His work on the wave nature of the electron has paralleled that of Dr. Davisson and confirmed the results.

Dr. C. J. Davisson

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Physics Teachers of Western Pennsylvania

The Association of American Physics Teachers of Western Pennsylvania and Environs met at Carnegie Institute of Technology, Pittsburgh, Pennsylvania, Saturday, November 6. The following program was presented:

1. **Two Demonstrations in Acoustics,** by CHARLES WILLIAMSON, *Carnegie Institute of Technology.*
2. **An Infrared Laboratory,** by C. E. LEBERKNIGHT, *Carnegie Institute of Technology.*
3. **More on Physics Definitions and Units,** by H. W. HARMON, *Grove City College.*
4. **Standards in Grading, with Particular Reference to the Curve of Normal Distribution,** by GEORGE E. DAVIS, *Duquesne University.*
5. **The Earth's Electric Field and Some Related Phenomena,** by M. C. HOLMES, *West Virginia University.*
6. **The Mechanism of Energy Loss in Magnetic Hysteresis,** by G. A. SCOTT, *University of Pittsburgh.*
7. **Lectures in Elementary Physics in the Small College,** by JAMES A. SWINDLER AND JOHN G. MOOREHEAD, *Westminster College.*
8. **A general topic: Training of Physics Students for Industry and Teaching** *suggested by* W. H. MICHENER.

Optical Society of America

The pleasant surroundings of the Lake Placid Club with its opportunities for recreation and informal discussions made the recent meeting of the Optical Society of America especially enjoyable and profitable to those who attended.

At an informal social gathering on Thursday evening kodachrome lantern slides and kodachrome motion pictures of the recent National Geographic-U. S. Navy Eclipse Expedition to Canton Island, taken by I. C. Gardner, Charles Bittinger and F. K. Richtmyer, members of the expedition, were shown. These included views of the encampment on the island and of the observing instruments as well as black and white and colored motion pictures of the eclipse itself. In addition to the Society and its guests the pictures attracted a large number of guests of the club who were not registered for the meeting of the Society. As an additional feature Loyd A. Jones showed a series of remarkable kodachrome motion pictures of growing organic crystals taken in polarized light, which had been awarded a medal at the annual exhibit of the Royal Photographic Society in London.

The symposium on optical materials Friday morning was unusually interesting and aroused much discussion. The following papers were presented in the symposium.

Optics and the Glass Industry. DR. E. D. TILLYER, American Optical Company
The Availability of Optical Glasses. DR. GEORGE W. MOREY, Geophysical Laboratory
Glass Requirements of the Optical Manufacturer. DR. W. B. RAYTON, Bausch & Lomb Optical Co.
Optical Glass at the National Bureau of Standards. DR. A. N. FINN, National Bureau of Standards
Crystal-Growing for Optical Purposes. PROFESSOR DONALD C. STOCKBARGER, Massachusetts Institute of Technology.

The feature of the annual dinner Friday evening was the award of the Frederic Ives Medal to Dr. Herbert E. Ives of the Bell Telephone Laboratories.

In accepting the medal Dr. Ives recalled incidents in the life of his father in whose honor the medal was endowed, and exhibited some of the early lantern slides which his father had made, using the original three-color mixing projection lantern constructed to exhibit them in natural colors.

At the dinner President Hardy introduced the newly elected officers of the Society:

President, PROFESSOR R. C. GIBBS, Cornell University
Vice President, DR. K. S. GIBSON, National Bureau of Standards
Members at large of the Board of Directors
H. P. GAGE, Corning Glass Works
WILLIAM F. MEGGERS, National Bureau of Standards
BRIAN O'BRIEN, University of Rochester

Fifty-seven members and thirty-five guests were registered at the meeting.

Society of Rheology

The Society of Rheology held its 9th annual meeting at Akron, Ohio on Friday and Saturday, October 22 and 23. The mornings of both days were devoted to technical sessions at which some sixteen papers were presented. Theoretical papers predominated, both in the general session held on Friday, and in Saturday's session which was devoted largely to the rheology of rubber.

On Friday afternoon members of the society were guests of the Guggenheim Airship Institute and the B. F. Goodrich Rubber Company; interesting inspection trips being provided by both organizations. The annual dinner of the society was held on Friday evening, and was followed by a demonstration of stress phenomena in glass, by H. R. Lillie, of the Corning Glass Company. This demonstration, which included moving pictures of strain measurements in polarized light, provided a vivid explanation of the relation between the strength of glass and its condition of internal strain.

At the business meeting of the society, held on Saturday afternoon, important changes were made in the society's membership and publication policies. The Rheology Leaflet, which was started in 1937 on an experimental and tentative basis, was scheduled for regular publication as a quarterly journal, to furnish not only news of the society but also abstracts and reviews of rheological literature. In line with this change, provision was made for associate members, paying annual dues of $2.00, to receive the Rheology Leaflet. Regular members will continue to receive both the Leaflet and the *Journal of Applied Physics*. As before, original papers presented to the society will be submitted for publication in the *Journal of Applied Physics*, while the Leaflet will serve as a medium for other literature and news relating to rheology.

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Television Tubes Now Available

Two cathode-ray tubes suitable for television reception are being made available to radio amateurs, educational institutions, laboratories and others interested in experimental television by the RCA Manufacturing Company. The tubes are both of the electromagnetic-deflection type and employ viewing screens on which the picture appears clearly, with a yellowish hue. They are known as RCA-1801, "Kinescopes" the former being a nine-inch tube and the latter a five-inch tube. They carry suggested list prices of $60 and $40, respectively. The "Kinescopes" each employ an electron gun and a fluorescent screen assembled within a vacuum tube. The negative electrode delivers a stream of electrons varying in intensity with the strength of the signals received. By means of magnetic deflection coils, this beam is made to scan the fluorescent screen which then emits light in proportion to the beam intensity. The beam can be made to trace a pattern of 441 lines, 30 times a second, giving picture definition substantially equivalent to a good photographic enlargement. The tubes will permit experimenters to build receivers to pick up experimental television transmissions.

Laboratories' Association

Representatives of twenty of the principal commercial laboratories of the country, at a meeting in Chicago last week, completed the organization of the American Council of Commercial Laboratories. One of the purposes of the Council will be the promotion of the proper use of scientific testing methods for the protection and certification of quality in advertised goods. In behalf of the Council it is stated that the members have been and will be carefully selected to include only organizations to which producers, retailers and consumers may look for unbiased determinations of quality. Engaging in research and testing for fees, these independent laboratories ascertain and report facts for clients and are uninfluenced by any ulterior consideration.

Laboratories which are adjuncts of other enterprises, or which are not self-supporting, or which, for any reason are not independent, are ineligible for membership in this council. The following officers and members of an executive committee were elected to serve during 1938: President—Preston S. Millar, Electrical Testing Laboratories, New York, New York. Vice President—Monroe L. Patzig, Patzig Testing Laboratories, Des Moines, Iowa; Secretary—D. E. Douty, U. S. Testing Company, Hoboken, New Jersey; Treasurer—A. R. Ellis, Pittsburgh Testing Laboratory Pittsburgh, Pennsylvania.

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Commercial Magnesium

It was announced at the New York Meeting of the Electrochemical Society that commercial production of magnesium can be carried on by a new "electrothermic" process which will make it as cheap as, or cheaper than, aluminum. In the process magnesite, which is a carbonite, is burnt to an oxide. The oxide is then heated in an electric furnace to approximately 2300°C and mixed with carbon in the form of coal or coke. In this step magnesium vapor and carbon monoxide gas are formed. The vapor is cooled by hydrogen gas, and magnesium powder is produced and is then redistilled to a metal of high purity.

★

New Type Telephones

A new type telephone set has recently been patented by two Bell Telephone Laboratories' engineers in which the bell and other necessary operating equipment is built into a unit not much larger than the present hand set. The use of this telephone set will eliminate the necessity of a separate bell box and will decrease considerably the time of installation.

★

Professor The Svedberg, University of Upsala, Sweden, Nobel Prize Winner in Chemistry for 1926, gave three public lectures at Carnegie Institute of Technology, Schenley Park, Pittsburgh, Pennsylvania, on "Molecular Migration under the Influence of Centrifugal, Osmotic and Electrical Forces" including "The Experimental Technique," "Investigations on Proteins," and "Investigations on Other Systems."

New Ultra-Sensitive d.c. Meter Announced by RCA

A new ultra-sensitive d.c. meter has been developed by the RCA electronic research laboratory. It is being made by the Engineering Products Division of the RCA Manufacturing Company. The meter was developed for accurate measurements of ionic and electronic currents, employing a new electronic circuit which operates with unusual stability and accuracy. It cannot be easily damaged or burned out by overload currents.

In current measurements the new meter provides for twelve scale ranges for measurement to 10,000 microamperes, the lowest full scale reading being 0.02 microamperes. For voltage measurements, eight other scale ranges are provided, from 0.1 volt to 500 volts, with a meter resistance of 5 megohms. For resistance measurements, two scale ranges are provided for measurements of from 0.1 to 100 megohms, and from 20 to 1000 megohms, with less than 0.5 volt across resistance. With 90 volts in series, up to 200,000 megohms can be measured. Conversion of the meter for the three forms of operation is accomplished by means of a selector switch. The ultra-sensitive d.c. meter is a self-contained, battery operated precision instrument, utilizing three RCA 1B-4 tubes. No external resistances or shunts are required.

★

Symposium on Wear of Metals

A reprint of the papers from the Symposium on "Wear of Metals" has recently been published by the American Society for Testing Materials. The papers included in this reprint are: Considerations Involved in the Wear Testing of Metals, by H. W. Gillett; Some Important Variables Encountered in Wear Tests on Cast Iron, by D. E. Ackerman; Wear of Metals from the Automotive Viewpoint, by W. E. Jominy; Wear from the Power Equipment Viewpoint, by N. L. Mochel; Wear of Metals in the Textile Industry, by A. Palmer; and Wear of Metals from the Railroad Viewpoint, by L. W. Wallace. Discussions of the papers are included and copies are available from the A. S. T. M. Headquarters, 260 South Broad Street, Philadelphia, at $1.25 each.

New Booklets Received

Current Titles

A new periodical entitled *Current Titles* has just been issued in which the titles of technical journals in the fields of engineering, technology, geology, physics and chemistry are listed. In the October issue there are 127 periodicals listed. This periodical is published monthly by Current Titles from Engineering Journals, 928 Broadway, New York City.

The Lovibond Comparator. This booklet consists of about 30 pages describing the comparator for colorimetric determinators made by the Kintometric, Ltd., Milford, Salisbury, England. It describes the uses of this instrument in many different fields.

Four Hundred Practical Applications for Monel Nickel and Inconel under Corrosive Conditions. The various applications of monel nickel and Inconel are described in a convenient form in this 26 page booklet. This booklet is published by the International Nickel Company, Inc., 67 Wall Street, New York City.

The General Radio Experimenter (8 pp). Volume XII, No. 6, November, 1937. This issue contains articles on radiofrequency source for the laboratory, adjustable attenuator, and improvements in the standard signal generator.

Among Our Contributors

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Contributed Original Research

Some Physical and Radiographic Properties of Metallic Intensifying Screens*

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The photographic intensifying action of metal screens has been known for a number of years but it is only with the advent of high voltages and thick materials of industrial radiography that their use has become especially advantageous. Lead is the material now commonly employed. The present work is an attempt to extend our quantitative knowledge of this subject by studying the various phenomena separately. The higher atomic number elements are shown to be the most suitable for intensifying screens and of these lead provides the best combination of properties. The front screen is more effective in its intensification than the back screen. This is explained on the basis of present-day knowledge of electron emission under x-ray excitation. Most of the intensifying action seems to be due to the electrons emitted and only a small part to characteristic and scattered x-rays. The intensification factor increases with kilovoltage and filtration between 160 and 200 kv and from 0.6 to 2.5 cm of steel, but apparently with sufficient decrease in x-ray wave-length, the intensification of lead foil decreases, because it is much less in gamma-ray radiography. The effect on intensification of variation in thickness of lead screens is very slight but, for practical reasons, it is desirable to have the back screen fairly thick. The thickness of the front screen is determined largely by its reduction of secondary radiation and a value from 0.010 to 0.015 cm is satisfactory. The experiments indicate that, in the reduction of secondary radiation by lead screens, differential *intensification* of the primary and secondary radiation is of major importance. Differential *absorption* by the front screen is also a factor. Lead screens are of advantage in welding and casting inspection but one of their most striking properties is shown in the radiography of small objects that normally require masking in which masking may often be eliminated because of the characteristics of the lead screens. The screens should be kept clean, since light materials absorb the intensifying radiation considerably. Visually, definition with the lead screen is just as good as it is with a nonscreen exposure but intimate contact in the cassette is essential. There seems to be little hope of making important improvements in the radiographic properties of lead screen, as e.g., by the addition of a fluorescing surface layer.

INTRODUCTION

THE fact that metal foils in contact with the emulsion intensify the photographic latent image during x-ray exposure has been known for a number of years.** Lomon[1] gave a good qualitative discussion of the practical results of this action in which he stated that lead and gold are the best metals because the intensification is greater the higher the atomic weight. A reduction in the amount of secondary radiation and an increase in the intensification with subject thickness penetrated were also noted. He referred to the earlier work of Chanoz[2] but this was undoubtedly due to a misinterpretation of Chanoz' experiments which demonstrated photographic *reversal* with x-rays. Steuart[3] reported excellent results with a back foil screen of silver-plated copper. Hodgson[4] tried several metals and found silver better than platinum. Schocken and Grebe[5] tried a few experiments with aluminum, copper, and lead foil screens operating as high as 160 kv with 1.5 mm of copper filtration and doubled the

* Communication No. 639 from the Kodak Research Laboratories.

** Since this paper was written the author has found brief mention of the intensifying action of metallic screens in the following references: W. C. Roentgen, Science **3**, 227 (1896). (Translated by A. Stanton.) J. T. Pitkin, Arch. Electrology and Radiol. **4**, 436 (1904).

[1] A. Lomon, J. de radiol. et d'électrol. **1**, 129 (1914–15).

[2] M. Chanoz, Lyon Médical, **110**, 616 (1908).

[3] W. Steuart, Archives of the Roentgen Ray **17**, 412 (1912–13).

[4] M. B. Hodgson, Phys. Rev. **12**, 431 (1918).

[5] K. Schocken and L. Grebe, Phot. Korr. **67**, 21, Heft 8 (1931).

TABLE I.

ELEMENT OR ALLOY	ATOMIC NUMBER	THICKNESS IN CM	REMARKS
Mg	12	.007	Courtesy of Aluminum Co. of America
Al	13	.0065	Baker & Co. >99% pure
Fe	26	.0065	Commercial steel shim stock
Ni	28	.006	Baker & Co. >99% pure
"Monel"	28.1	.007	Baker & Co., 68% Ni, 29% Cu, 1.5% Fe, 1.1% Mn
Cu	29	.005	Commercial, electrolytic
Zn	30	.006	Baker & Co. >99.9% pure
Mo	42	.006	Baker & Co. >99.9% pure
Ag	47	.0055	Baker & Co. >99% pure
Cd	48	.006	Baker & Co. >99.9% pure
Sn	50	.0045	Commercial foil, Will Corporation
"No. 417"	60.7	.006	Baker & Co., 60% Au, 40% Pd >99% pure
"Solder 5"	61.6	.007	Commercial 50% Sn, 50% Pb
"Solder 3"	67.2	.0065	Commercial 33% Sn, 67% Pb
Ta	73	.006	Courtesy of Fansteel Metallurgical Corp.
W	74	.005–.006	Courtesy of General Electric Co.
Pt	78	.005	Rolled by H. A. Swift, Rochester, N. Y.
Au	79	.0065	Baker & Co. >99% pure
Pb	82	.005	Baker & Co. >99.9% pure
Bi	83	.004	Laboratory stock, rolled by Baker & Co.

speed of the photographic film with the lead screen. Aluminum and copper were less effective, the action of all three being in the order of atomic weight. They concluded that most of the intensification was due to electron action because it was so strongly absorbed in celluloid. In spite of early reports[3] that metal screens were effective in medical radiography, present-day tests indicate that in this type of radiography they give no important intensification. Perhaps this is due to differences between the earlier and more recent photographic materials.

Lead screens are used to some extent at the present time in industrial radiography, especially when gamma-rays are used to penetrate thick sections of steel.[6–8] Intensification, removal of some of the secondary radiation, increase in film contrast, and maintenance of good definition are all effects of worthwhile magnitude.

Since the brief work by Schocken and Grebe seems to be the only quantitative information on this subject, it seemed desirable to make a new approach to the problem, studying the different phenomena separately and to determine, by a more extensive survey of materials, which is the most effective as an intensifying screen, and then to study the optimum conditions for its use. The investigation of a fairly large number of materials was justified since it was recognized that the intensifying action must be due to more than one effect, and it would, therefore, be difficult, even from our present knowledge of x-rays, to predict the total resultant effect.

PHYSICAL EXPERIMENTS

Table I gives a list of the materials studied and some of the data pertaining to them. "Effective" atomic numbers of the alloys were computed from the relation

$$Z_e=\frac{(f_1/A_1)Z_1+(f_2/A_2)Z_2+\cdots(f_n/A_n)Z_n}{(f_1/A_1)+(f_2/A_2)+\cdots(f_n/A_n)},$$

where Z_e is the effective atomic number, f_n is the proportion by weight of element n in the alloy; A_n and Z_n are atomic weight and atomic number, respectively. This method of averaging assumes that the dependent variable is a linear function of Z. Narrow strips, in groups of five, were cemented to pieces of cardboard, leaving one area blank for the direct x-ray exposure alone. When clamped in a cassette in contact with the emulsion of *single coated* Eastman x-ray film, they were placed in a sensitometer (Fig. 1) and moved across the x-ray beam in a geometrical time series

[6] C. S. Barrett, R. A. Gezelius, and R. F. Mehl, Metals and Alloys, **1**, 872 (1930).
[7] G. L. Clark, *Applied X-Rays*, second edition (McGraw-Hill Book Co., 1932), p. 116.
[8] J. T. Norton, in *Symposium on Radiography and X-Ray Diffraction*, (Am. Soc. for Testing Materials, Philadelphia, 1936), p. 8.

of steps varying from $\frac{1}{8}$ to 4 minutes. This motion was remotely controlled electromagnetically from the operator's booth outside the x-ray room. The strips were processed under carefully controlled conditions to obtain the highest accuracy for this type of work. Details are not presented here because the subject has been fully discussed by Wilsey.[9] Density readings were made on a Martens polarization photometer. The x-ray tube was operated at 200 kv constant potential, 4 ma,

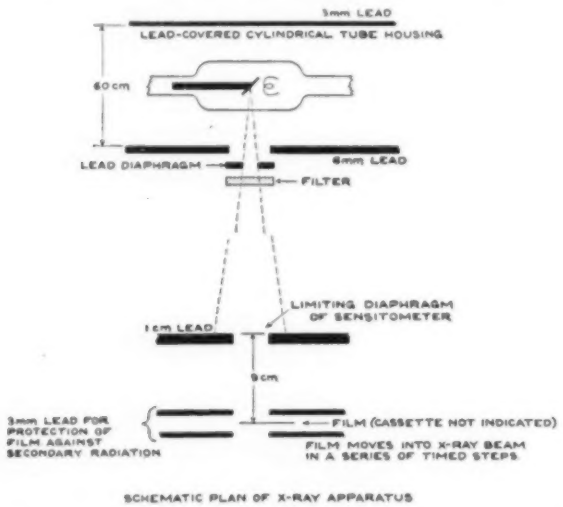

FIG. 1. A series of known exposures may be made upon the film with the sensitometer to determine the response of the film to the radiation. X-ray photographic intensities may also be compared. For a single exposure the sensitometer need not be used, since the exposure may be timed by a lead shutter near the filter.

0.95 cm cold rolled steel filtration, and 100 cm target-film distance. Secondary radiation from the filter was found to be about 3 percent of the total intensity.[10] The term "constant potential" is based on full wave rectification with a capacity of 0.025 microfarad across the high tension line giving by *approximate calculation* a ripple of 330 volts per ma. For the same radiographic results on this machine without condensers the peak voltage is increased about 10 percent. Voltage calibration was effected using Taylor-Shallcross[11] high voltage shielded resistors as a voltmeter multiplier. All factors considered, the voltage is probably known to within 2 percent for a given machine setting but the maintenance of constancy of voltage is more precise than this.

Data were taken with the foils used first as *back* intensifying screens, that is, with the film on the tube side of the screen, and then as *front* intensifying screens. Fig. 2 gives the results of these measurements. Each datum is the average of 3 sensitometric runs. The ordinates are determined from the reciprocal of the time of exposure required to produce a density of 1.00. The justification for a direct interpretation of reciprocal time as proportional to intensity is based on the work of G. E. Bell,[12] and A. Becker and E. Kipphan,[13] who show that the reciprocity relation, namely, intensity $\times$ time = a constant, holds for direct x-ray and cathode-ray exposures. The ordinate value 1.00 in Fig. 2 represents the exposure due to direct x-rays and, therefore, a full ordinate minus 1.00 is the intensifying action of the screen. The x-ray intensities for front screen exposures are not quite as great as those for back screen exposures because of x-ray absorption in the front screen, but this is very small as will be shown later. In spite of this, the action of the front screen is definitely greater than the action of the back screen.

At first it seemed that the observational error among the high atomic number elements was very large, particularly for the back screen data, but examination of the separate runs indicated a systematic effect which justified averaging the data for each material and plotting the lower curve of Fig. 3. In a simple theory to account for electron emission from the surfaces of solids irradiated with x-rays, Holthusen[14] assumed that the total electron energy would be (1) proportional to the mass absorption coefficient μ_m, (2) inversely proportional to the mass absorption coefficient of the electrons in the parent substance, and (3) proportional to the difference between the quantum energy of the exciting radiation and the quantum energy corresponding to the K absorption discontinuity. The electron intensity would therefore be proportional to $\mu_m(1/\lambda - 1/\lambda\kappa)$, where $(1/\lambda - 1/\lambda\kappa)$ corresponds to assumption (3). Assumption (2) does not intro-

[9] R. B. Wilsey, Am. J. Roentgenol. **32**, 789 (1934).
[10] H. E. Seemann, "Secondary Radiation Intensity as a Function of Certain Geometrical Variables," to be published in Am. J. Roentgenol.
[11] L. S. Taylor, Nat. Bur. Standards J. Research, **5**, 609 (1930) (Research Paper No. 217).
[12] G. E. Bell, Brit. J. Radiol. **9**, 578 (1936).
[13] A. Becker and E. Kipphan, Ann. d. Physik **10**, 15 (1931).
[14] H. Holthusen, Ergebnisse der Medizinischen Strahlenforschung **1**, 383 (1925).

duce a variable since the mass absorption coefficient of electrons is taken as essentially constant for all materials. The Compton effect is not considered. Graphical illustration of Holthusen's theory is given in Fig. 3 for three wave-lengths which occur in the 200 kilovolt x-ray spectrum. The theory seems to account qualitatively for the experimental results which are shown later to be due principally to electron action. The shift of the peak toward lower atomic numbers with increase in wave-length is verified experimentally by the fact that of the elements copper, silver, tin, tantalum, and lead tested as back screens for medical diagnostic qualities of x-radiation, silver and tin gave the greatest intensification. The total amount, however, represented a radiographic speed gain of only about 25 percent and this was lost in front screen absorption when double screens were used.

While the *exact* location of experimental data must not be overemphasized because of normal observational errors, it is felt that the experimental evidence is against the use of the equation given above for calculating the effective atomic number of an alloy when the constituents differ very widely in atomic number. This equation

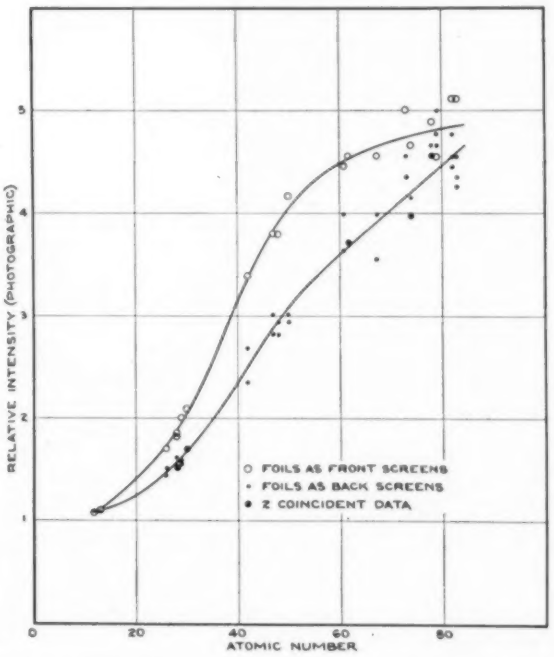

FIG. 2. Relative photographic x-ray intensities as a function of the atomic number of the screen material.

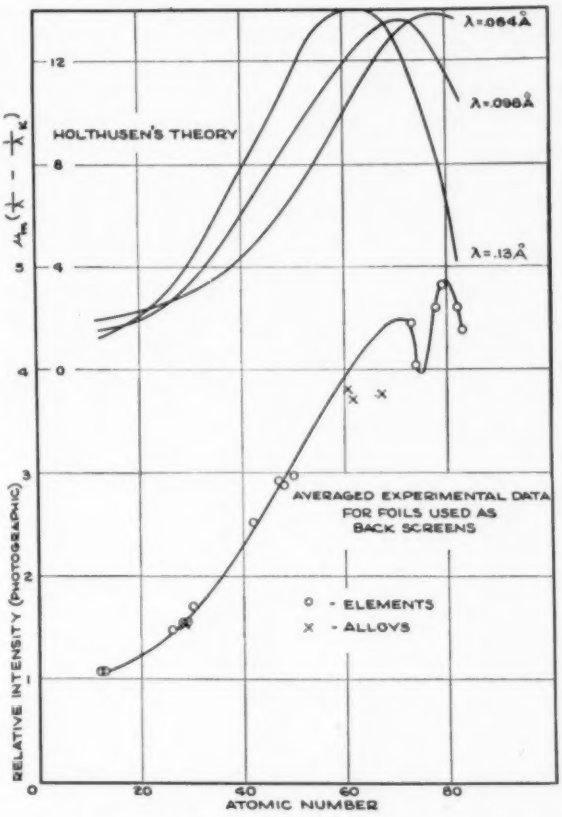

FIG. 3. Results for back screens qualitatively explained by Holthusen's theory. The variation of mass absorption coefficient for electrons of different velocities has been neglected in the calculated curves.

assumes a linear relationship between atomic number and the property considered, whereas the theoretical curves (Fig. 3) show that a discrepancy would be expected.*

An analysis of the intensifying "radiation" from lead foil will now be made. It is expected, of course, that part of this will be electrons and part secondary x-rays. A method of distinguishing between electron action and secondary x-ray action is made possible by the difference in the absorption laws for these two. It has been found[15] that all materials absorb electrons with *approximate* equality for the same mass penetrated, and atomic number has little effect. X-ray absorption,

* If two strips of film are crossed with their emulsion sides in contact and exposed to hard radiation, it will be found that they intensify each other to some extent. Also, a developed photographic negative will produce *another negative* if used as an intensifying screen. The response is very weak but shows that when the areal concentration of an element is *small*, the intensification depends on the amount per unit area.

[15] P. Lenard and A. Becker, "Kathodenstrahlen," *Handbuch der Experimental Physik*, Vol. 14 (1927), p. 183.

on the other hand, is very markedly determined by atomic number for the same mass of material penetrated. Accordingly, stepped wedges of thin aluminum and Kodapak† were placed between a 0.007 cm lead screen and the x-ray film which, in this case, was the regular double-coated type. The aluminum tablet was made up of foil of 0.00436 gram per square centimeter. The Kodapak weighed 0.00295 gram per square centimeter. Three 2-minute exposures at 200 kv, 4 ma, 100 cm target-film distance with 1.27 cm of cold rolled steel filter were made and the results averaged. The x-ray beam was turned on and off by a lead shutter to avoid the variations associated with starting both the x-ray machine and exposure at the same time. The densities were evaluated as photographic intensities from the appropriate characteristic curves run on the sensitometer.

Referring to Fig. 4, the relative intensities transmitted by the various steps of the tablets are shown plotted as a function of the mass per square centimeter. An intensity of 100 represents the exposure due to direct x-rays and lead-screen intensification with no absorbing material interposed. Within the limits of experimental error, the agreement between aluminum and Kodapak

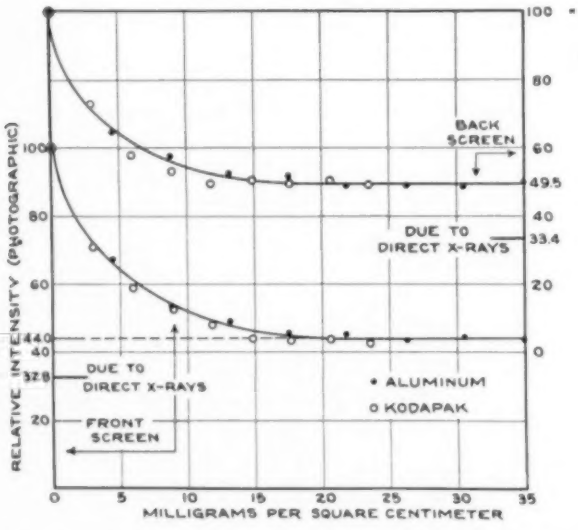

FIG. 4. Electron absorption curves in aluminum and Kodapak.

† "Kodapak" is the trade name of a cellulose acetate wrapping material containing some plasticizer and manufactured by the Eastman Kodak Co. Its specific gravity is about 1.3. It has a mass absorption coefficient about 1/5 that of aluminum in the 1A region.

absorption is quite satisfactory. The horizontal portion of the curves is interpreted as indicating complete electron absorption, but there is still a certain amount of radiation penetrating the thickest portions of the tablets. This must be the characteristic K radiation of lead and the scattered radiation both of which are only slightly absorbed. The wave-length limits of the L series of lead (0.84A to 1.35A) are appreciably absorbed in aluminum of the thicknesses used here, but the evidence given by these curves indicates that the total L series intensity must be quite small compared with the other effects for, if it were not, the curves would continue their downward trend. Of course, interpreting the L series as a very low intensity simply refers to the intensity being of the order of magnitude of the experimental error. The M series is almost completely absorbed by the thinnest step. It may be seen from these curves that the electron intensity is 84 percent of the *intensifying action* for the front screen and 75 percent for the back screen. The intensity of the *direct* x-rays is about 33 percent of the *total* in both cases. This large percentage of the total results from the use of only one screen with *double-coated* film. The shape of these curves differs from the electron absorption curves of Schonland,[16] which are almost linear, in just the manner to be expected from the fact that the x-ray beam is not homogeneous and, therefore, will not give a homogeneous velocity to the electrons. Furthermore, even if the electrons from the lead atoms were all from outer shells and due to 200 kv x-rays, there would be a wide range of velocities among those emitted because of partial absorption of electrons from various depths of the metal. This may account for the fact that complete electron absorption occurs at about 20 milligrams per square centimeter of aluminum, while interpolation of Schonland's[16] and Varder's[17] results gives a velocity corresponding to 135 kilovolts for electrons thus absorbed. Probably very few actually are 200 kilovolt electrons. Assuming that the mass absorption coefficient of all materials for electrons is the same, it may be concluded that no electrons come from more than 0.0018 centimeter below

[16] B. F. J. Schonland, Proc. Roy. Soc. **108**, 187 (1925); M. Allen, J. Frank. Inst. **214**, 25 (1932).
[17] R. W. Varder, Phil. Mag. **29**, 725 (1915).

the surface of the lead for the radiation quality used here. The ratio of electrons emitted in the forward direction from the front screen to those emitted in the backward direction by the back screen is 1.12, which agrees reasonably well with the results quoted by Compton and Allison.[18]

It is of interest to analyze Fig. 2 in this connection. As explained before, the intensity above ordinate value 1.00 represents that due to the intensification from the metal screens. From the data available, this cannot be separated into its component radiations, but an approximate relation between forward and backward emitted electrons may be obtained by taking the ratio of the intensifying action of the front and back screens as a function of atomic number. These values from the graphs of Fig. 2 show a decrease in the ratio with increase in atomic number. This would be expected from the work of Woo[19] on the variation in prominence of the unmodified line with atomic number. Also, Bragg and Porter's[18] results on the emergent-incident ratio of emitted electrons as a function of atomic number show this decrease. It seems, therefore, that the curves of Fig. 4 can be interpreted as electron absorption curves.

Radiographic Characteristics of Lead

After a short study had been made of the physical aspects of metallic intensifying screens in which lead was found to be the most suitable material, some of the properties of this screen were then studied with their application more definitely in mind. It is natural to raise such questions as "How does the intensification change with the kilovoltage or with the filtration?" "Does the thickness of the screen make any difference?" "To what extent does the screen remove secondary radiation?" etc.

The intensification factor of any kind of radiographic intensifying screen is defined as the ratio of exposure without screen to the exposure with the screen which will produce some specified density. On the basis of this definition, the ordinate in Fig. 2 could be labeled "intensification factor," but since the specimens of foil were not used as double screens, which would be the practical method, the designation used seemed more desirable. Fig. 5 shows the variation of the intensification factor with kilovoltage. The data given were not direct observations but were derived from the graphical averaging of the log exposure-log kilovoltage results which are straight lines. The intensification factor is determined for a density of 1.00 throughout this work since this is approximately the density region desirable in industrial radiographs. It may be noted that the graph of Fig. 5 is not quite a straight line but is slightly concave toward the voltage axis. Fig. 6 shows the variation of intensification factor with steel filter thickness at 200 kv constant potential. The increase in intensification factor with kilovoltage and steel filter thickness may be interpreted as different aspects of the same thing, namely, that the shorter the wave-length regions, the greater is the intensification factor. Since both curves are slightly concave toward their axes of abscissae, it may be expected that the intensification factor will somewhere reach a maximum and then perhaps decrease. There is some evidence for this, inasmuch as the intensification factor with gamma-rays penetrating thick sections of steel is only about 1.3.[8] This phenomenon of increase of intensification factor with decreasing wave-length, and then reversal of the trend, is a well-recognized property of the commercial calcium tungstate intensifying screen.

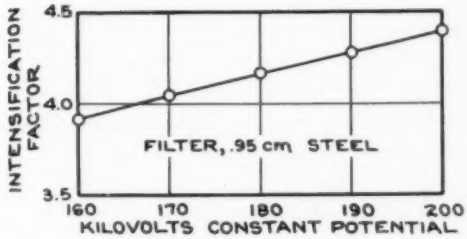

Fig. 5. Intensification factor for double lead screens as a function of kilovoltage with constant filtration.

Figure 7 shows the effect on the photographic response of variation of thickness of front and back screens. Observations for the front screen were made at 200 kv constant potential and 1.27 cm steel filtration. Those for the back screen were made similarly but with 1.9 cm of steel. They are, therefore, not strictly comparable but illustrative. As far as the intensification is concerned, there is very little to be gained in having a back screen any thicker than that which can be

[18] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Co., 1935) p. 564.
[19] Quoted by Compton and Allison, p. 205.

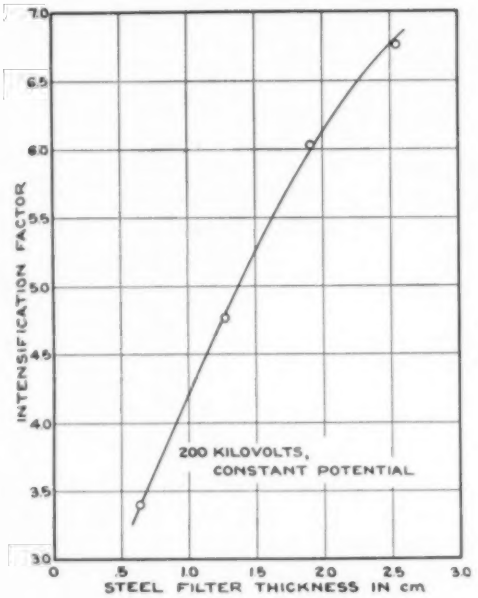

FIG. 6. Intensification factor for double lead screens as a function of filter thickness for constant kilovoltage. Beginning with the thinnest steel, the effective wave-lengths are 0.117, 0.102, 0.094 and 0.085A, respectively. These were determined from the tangents to the absorption curve.

conveniently handled. Since electron absorption is very high, the slight increase in intensity due to increased thickness of the back screen is interpreted as additional secondary x-radiation. The characteristic which determines the optimum thickness of the front screen will be discussed later. The intensity-thickness graph for the front screen is a straight line. Since the *true absorption* curve would be convex toward the thickness axis, this experimental result is interpreted as the result of the addition of secondary radiation.

It is well known by those who use lead screens that they improve the quality of the radiograph by removing some of the secondary radiation which is nonimage-forming and which decreases subject contrast in the same way as an accidental fogging of film. It seemed of interest to make a few tests to learn a little more about secondary radiation removal. A simple method has been worked out[10] which makes it possible to distinguish experimentally between the direct radiation from the x-ray tube and the secondary radiation contributed by a specimen of uniform thickness and composition. Fig. 8 shows the results of the application of this method to the study of secondary radiation in the radiography of a piece of aluminum 5.1 cm thick at 150 kv and a piece of cold rolled steel 1.27 cm thick at 200 kv. In the preceding experiments, the steel was used as a filter, that is, secondary radiation was reduced to a small percentage by having the filter well diaphragmed and near the tube. In practical radiography, of course, the specimen must be as near the film as possible in order to have good definition of flaws or other details. This means however, that a large amount of secondary radiation will be recorded by the film. Since the secondary radiation is, in general, of a longer wave-length than the primary radiation, it would be expected that a lead filter between specimen and film would preferentially absorb it. The ratio of secondary radiation intensity I_S to direct radiation intensity I_D is here plotted as a function of the front lead screen thickness. The secondary radiation from the aluminum is about 3.5 times as intense as the direct radiation without screens but it falls off with the use of a back screen and an increase in front screen thickness. For the steel, the secondary radiation is about 1.6 times as intense as the direct beam and changes in about the same proportion with the screen thickness as does the aluminum. It may be somewhat surprising to note that the reduction of secondary radiation is not primarily an absorption phenomenon, for the use of a back screen *without* the front screen exerts a very material effect in diminishing secondary radiation. Preliminary experiments had shown that a front lead screen with heavy cardboard between it and the film did not do a great deal of good in secondary radiation removal but the experiments

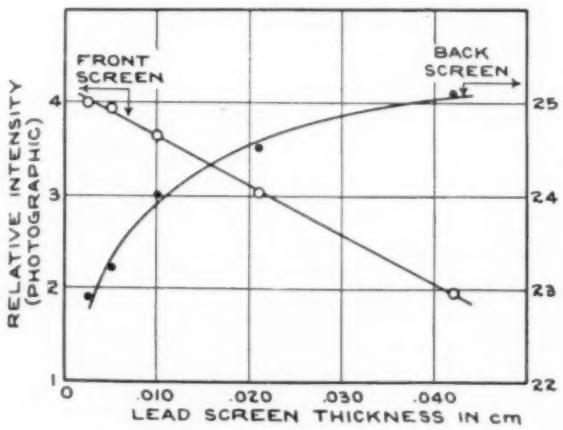

FIG. 7. Relative photographic intensity as a function of screen thickness. The two ordinates are not on the same scale.

were not carried further because mounting of the lead in this manner still permitted some intensification, as illustrated in Fig. 4, so that it could not be considered strictly as an absorber. It seemed preferable to illustrate the principle by using a back screen alone and then obtaining the rest of the data with the front screen thickness as a variable, the back screen thickness remaining constant. The trend of the curves in Figs. 5 and 6 indicates the reason for the large reduction in secondary radiation produced by a back screen with front screen omitted, because the direct radiation, being of a shorter wave-length, is *intensified* more than the secondary radiation.* The curves of Fig. 8 show little advantage in reduction of secondary radiation from the use of front screen thicknesses in excess of 0.010 to 0.015 cm. Greater thicknesses will appreciably decrease the photographic intensity of the direct x-rays, as indicated in Fig. 7. Furthermore, additional material in the screen, other properties being neglected, is equivalent to the addition of thickness to the subject radiographed, tending to decrease the sensitivity to defects.

Tin screens 0.004 cm thick were found to reduce secondary radiation by about 25 percent under conditions similar to those of medical radiography at 80 kv peak with a phantom consisting of 5 cm of water plus 0.6 cm of Tenite. This reduction must be due primarily to differential absorption, since the intensification, as mentioned above, is very small. The result is similar to that obtained by Wilsey[20] using 0.0025 cm of lead foil only as a filter.

It has been mentioned in the literature that the contrast of the film, that is, the slope of the density-log exposure curve, is somewhat greater with the metal intensifying screens than without. This difference in the film characteristic has been noted in the experiments reported here but its magnitude is an increase of from 10 to 20 percent over the direct x-ray exposure. It is, therefore, definitely noticeable but not very pronounced. It was not detected in the data for Fig. 2 where a single screen and single-coated film were used. It is observed, however, with double lead screens and 1.2 cm steel filtration. It is not known definitely whether this change in contrast is due entirely to electron action, for a very slight increase in contrast has been noted in the characteristic curve taken with x-rays generated at about 17 kv and filtered with 0.015 cm of aluminum (and Slack window) giving an effective wave-length of about 1.2A, the region of the *L* radiation of lead. The film contrast for this radiation seemed to be intermediate between that for direct high voltage penetrating radiation and that for lead intensification. The differences are slight so that it is hard to analyze the results but it seems likely that the electrons are largely responsible for the increased contrast.

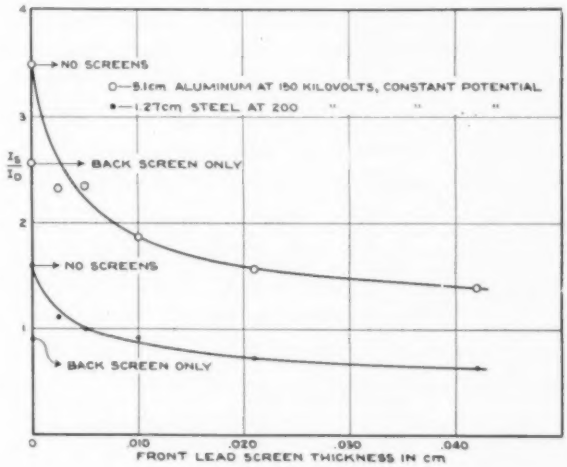

FIG. 8. Ratio of secondary radiation intensity to direct as a function of front screen thickness for two typical cases. A constant back screen thickness of 0.007 cm was used.

To those who have had experience in the practical radiography of industrial materials, the high intensification factors reported here for lead screens may be somewhat surprising. The apparent discrepancy between these results and practical tests is easily explained by comparison of Figs. 6 and 8. The given thicknesses of steel in the data of Fig. 6 were used as filters, that is, the secondary radiation from them was a very small fraction of the incident beam. In radiography, of course, a large amount of secondary radiation is present because of the close proximity of steel and film. Two partially compensating effects

* A difference in intensification of primary and secondary radiation probably exists for calcium tungstate screens but its importance is not known as no quantitative study has been made. The intensification factor increases with kilovoltage and filtration up to about 150 kv and 1.3 cm steel thickness but then begins to decrease. It is conceivable, therefore, that under some conditions the relative effect of secondary radiation may be reduced and under others increased with the use of calcium tungstate screens.

[20] R. B. Wilsey, Am. J. Roentgenol. **8**, 328 (1921).

exist. On the one hand, the lead screen gives a fairly high intensification. On the other hand, as indicated in Fig. 8, it removes considerable secondary radiation. Thus, the intensifying action is tending to increase the density in the radiograph, while the reduction of secondary radiation is tending to decrease it. It is quite conceivable, therefore, that in a given case, the intensification may be just compensated for by the removal of secondary radiation and, therefore, the time of exposure required to produce a certain density will be the same whether lead screens are used or not. In exposures through the heavier sections, a very material reduction in the time of exposure with lead screens will be noted but whether the exposure time is affected or not there will usually be an improvement in radiographic quality.

It has been shown in Fig. 7 that the advantage of a very thick back screen is practically negligible as far as direct radiographic results are concerned but, under practical operating conditions, there is good reason for having a fairly thick back screen to absorb radiation scattered from matter behind the film, since back-scattered radiation is sometimes intense enough to produce appreciable fogging of the film or even to record the spring clips on the *back* of the cassette. So much of the intensifying radiation comes from surface layers of the lead that the slight additional radiation, contributed by a thicker screen, is negligible in affecting definition. Visual comparisons of radiographs of definition test objects made with and without lead screens show no loss in definition. Intimate contact between screen and film must of course be maintained to insure good definition.

Lead screens should be kept as free as possible from dirt and loose particles for, as shown in Fig. 4, very light thin materials produce a large absorption of the intensifying radiation. An interesting illustration of this absorption is given by placing a piece of paper between film and screen and making an exposure at high voltage through a thick, uniform piece of steel. Close examination of the area covered by the paper will disclose a record of the paper structure. In other words, this is an "electron radiograph" of paper. Such a radiograph has been reported by Coolidge and Moore[21] using a cathode-ray beam outside the tube.

It is important that the foil used be free from any kind of *surface coating*, such as is sometimes used to give it a high gloss. During early preliminary studies of lead screens, some foil which had a very thin coating of tin was inadvertently used and the resulting radiographs were badly streaked. This was the result of nonuniformity in the thickness of the very thin tin coating so that essentially lead intensification was obtained in one area and tin intensification in the areas where the tin was thick. The use of small amounts of other elements to form a homogeneous alloy will have little effect on the radiographic properties of the screen and may make it harder and less likely to hold dirt than ordinary lead. A sharp distinction is therefore made between surface coatings of other materials and their use as small percentage ingredients of a homogeneous lead alloy.

While there is a marked improvement in the quality of a radiograph of a weld or casting with the lead screen technique as compared to the non-screen technique, one of the most striking advantages of this screen is in the radiography of small objects which do not cover the entire film and therefore need to be masked. The masking process is usually quite tedious but the use of a lead intensifying screen in many cases makes this unnecessary, for it acts primarily as a strong *absorber* of the intense radiation *around* the subject and, therefore, reduces the amount of overexposure in this region. Overexposure of the film at the *edge* of the image produces serious halation *in* the image as a result of secondary radiation from cassette, film, etc. If the absorption of the lead for the unfiltered radiation is inadequate, the addition of a thin steel or copper filter may suffice. The radiography of objects having a wide range of thicknesses is therefore facilitated with lead screens, since they tend to reduce the relative exposure and excessive secondary radiation from the thinner portions, at the same time increasing the exposure over the thicker portions. In other words, there is a general tendency toward reduction of *subject contrast* because of differential intensification

[21] W. D. Coolidge and C. N. Moore, Gen. Elec. Rev. **35**, 413 (1932).

simultaneously with increased contrast produced by removal of secondary radiation.

Lead foil has been successfully cemented to aluminum alloy sheets thus making a durable screen and one which is easily handled. Cementing to cardboard or cellulose acetate is probably satisfactory, though a metal backing has some advantages. Metal backed foil may be permanently bent to curvatures found in cylindrical vessels and used in a black envelope without a cassette, provided a curved form with sponge rubber facing can be wedged against the packet to insure good contact. The metal backed screen may be permanently fastened in conventional cassettes or used interchangeably with calcium tungstate screens.

In discussing the phenomenon of lead screen intensification with several persons, it was suggested that screen efficiency might be increased by coating it with a material which fluoresces under electron excitation. The *addition* of electron excitation from the lead to the normal excitation of the fluorescent material by the x-rays was considered a fruitful possibility. A simple experiment was tried to test this by using a layer of calcium tungstate as a front screen without its cardboard support. Part of this layer was backed with lead foil. The increase in density due to the lead backing was just detectable. When it is considered that the tungsten atoms *within* the calcium tungstate crystals emit electrons much the same as lead atoms, it is not so surprising that there is little to be gained from the lead. The slight gain noted may be the result of secondary x-rays, of course, and not of electrons. If some fluorescent material should prove to be suitable from the standpoint of speed, the problem of definition would then arise because of the finite thickness of the fluorescent layer. While it is impossible to predict, the chance that the conversion of electron energy to fluorescence and from fluorescence to latent image will be more efficient than the conversion directly from electron energy to latent image seems remote. Electrons are, of course, themselves very strongly absorbed in the silver bromide emulsion. Von Borries and Knoll[22] have reported that the difference in efficiency between the two methods of obtaining photographic density is not large. They found electrons above 20 kv to be more efficient photographically when used directly than when used with the "Schleede ZnS-Ag" screen as an intermediary.

[22] B. von Borries and M. Knoll, Physik. Zeits. **35**, 279 (1934).

A Characteristic of the Copper Arc During the Formative Period

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This paper gives the results of an investigation of the voltage characteristic of the copper arc in air during the process of arc formation. A pair of copper electrodes, initially in contact and carrying current, were separated and the voltage arising across the arc gap was studied. Cathode-ray oscillograms indicate that at the instant of contact separation there develops across the gap a voltage of about 12.2 volts. This voltage was attained in less than 10^{-5} seconds. This rapidly developed voltage was found to be independent of arc current for current values up to 100 amperes, which suggests that the rapidly developed initial voltage corresponds to the cathode fall of the arc.

WHEN an arc is initiated by the separation of a pair of contacts, a voltage arises practically instantaneously across the arc gap. The value of this rapidly developed voltage, at a break that occurs in the voltage characteristic of the arc as the arc is lengthened, is found to be reproducible and independent of the magnitude of the current flowing in the arc. This suggests that the rapidly developed arc voltage may be a measure of the cathode fall of the arc inasmuch as only the cathode fall is independent of arc current.

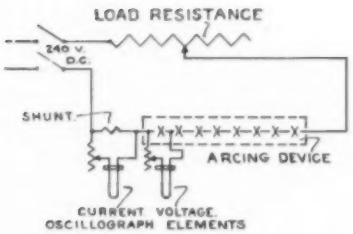

FIG. 1. Circuit using electromagnetic oscillograph.

In the initial experiments, current was caused to flow through a circuit which included a pair of copper contacts, which were initially in electrical contact. The contacts were separated, and the voltage arising across the arc gap was studied by means of an electromagnetic oscillograph and by means of a cathode-ray oscillograph in later experiments.

A diagram of the connections is shown in Fig. 1 for the circuit using the electromagnetic oscillograph. The power supply was 240 volts, d.c., and the current was limited by resistance to values between 15 and 105 amperes. An arcing device is indicated which consists of a number of arc gaps in series, the need for which is indicated by the following considerations. The arc voltage across a single gap is of the order of 20 volts, and is impressed on one of the oscillograph elements. It is desirable that the oscillograph be adjusted to give full deflection at this voltage, but, if only one arc were used, full line voltage (240 volts) would be impressed on the oscillograph element should the arc be interrupted, and this would damage the element. To prevent this, the arcing device indicated in Fig. 1 was used, which caused a plurality of arcs in series to be introduced into the circuit so that when the arcs are extinguished, the voltage across any one arc gap is zero. The arc gaps were introduced progressively, and the oscillograph was connected to record the voltage of one of the first gaps to open. The arcing device was operated automatically upon tripping the oscillograph, and a record was obtained of the voltage change across the gap under observation together with the arc current and a timing wave.

A typical oscillogram is shown in Fig. 2. The voltage trace is a trapezoid-like figure with an initial rise in voltage, E_1, that is practically instantaneous. Thereafter the voltage rises more gradually until the series arc gaps cause the current to be interrupted, and the arc voltage falls to zero. The initial arc voltage is designated E_1, and the voltage at current interruption is designated E_2. The oscillogram shows the decay of current in the circuit, and also gives a 62.5 cycle wave as a timing means.

In Fig. 2 the oscillograph element overtraveled before breaking away from the initial portion (E_1) of the trace to record the rising arc voltage due to the combined influence of decreasing arc current and increasing arc length. This overshooting of the oscillograph element was investigated and a correction factor was determined to be applied to the maximum initial deflection to give the true value of the initial arc voltage E_1.

The correction factor was obtained by making oscillograms of the sudden application of a known

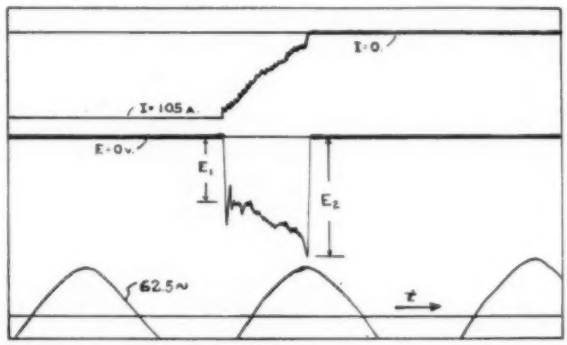

FIG. 2. Oscillogram (circuit Fig. 1). E_1 = initial arc voltage.

continuous voltage to the oscillograph element. The ratio of the steady deflection to the maximum deflection gives the correction factor. In Fig. 2 the correction factor was 0.745 and was determined with a calibrating voltage that was approximately equal to the initial voltage E_1 observed in the experiments.

EXPERIMENTAL

Typical data obtained with the electromagnetic oscillograph are given in Table I. The values of the initial arc voltage, E_1, range from 11.2 volts to 15.4 volts with an average value of 13.3 volts. Upon examination of the values of E_1 it is found that no correlation with current exists for arc currents up to 104 amperes. For example, the average value of E_1 for an arc current of 104 amperes is 13.7 volts. This value, however, is exceeded in the cases for arc currents of 14 and

TABLE I. *Applied voltage = 240 v, d.c. Copper contacts.*

Test No.	Osc. No.	Arc Current I (amp.)	E_1* (Volts)
1	B 349	14.	14.1
2	B 350	29.4	12.2
3	B 350	44.5	13.2
4	B 351	59.5	13.9
5	B 351	74.	13.0
6	B 352	88.5	12.6
7	B 352	104.	11.2
8	B 354	104.	15.4
9	B 354	104.	14.5
		Average	13.3

* Overshooting factor applied.

59.5 amperes, respectively. Other oscillograms made with an applied voltage of 120 volts d.c. gave values of E_1 similar to those obtained when the applied voltage was 240 volts d.c.

In subsequent experiments, use of the electromagnetic oscillograph was discontinued because of the inherent frequency limitation of the instrument which gave rise to doubts as to the actual value of the initial voltage E_1 and also as to the time for this voltage to attain its full value. A cathode-ray oscillograph was used instead of the electromagnetic oscillograph. This instrument was an RCA type TMV-122B and is equipped with a hot cathode-type tube and provides amplifiers to give a sensitivity of 2 volts per inch of deflection. The amplifiers are designed for alternating current only, and when applied to the problem at hand, made necessary a change in the method. A different type of arcing device was employed which consisted of a single pair of copper contacts operated by a motor-driven cam. A circuit diagram is given in Fig. 3. The arcing device was arranged so that the contacts were separated a short distance to form an arc and were then closed, without causing the arc current to be interrupted. This permitted high sensitivity to be employed without

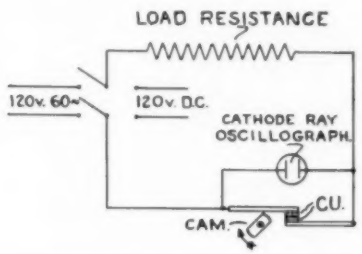

FIG. 3. Circuit using cathode-ray oscillograph.

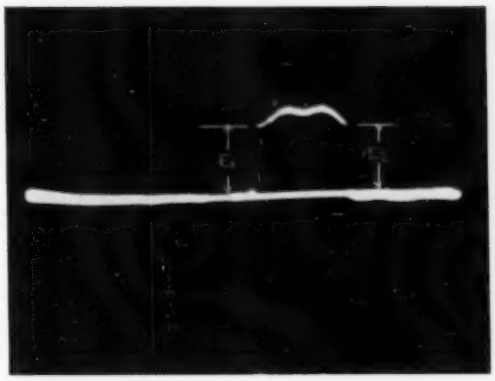

FIG. 4. Oscillogram (circuit Fig. 3). Arc voltage using direct current. E_1 = initial arc voltage.

full line voltage being applied to the oscillograph. Fig. 4 is a typical record obtained with the cathode-ray oscillograph in which the initial arc voltage is followed by a period during which the gap voltage increases and then decreases due to variations in the arc length, and then the contacts close, whereupon the voltage falls to zero.

In the experiments, a synchronous motor was used to operate the arc gap and caused the formation of the arc at a rate of 30 times per second, to which frequency or some multiple thereof the cathode-ray oscillograph was synchronized. Dividers were used to measure the deflection of the electron beam corresponding to the initial voltage E_1 and a voltage calibration was made of the deflection. In making the calibration, the calibrating voltage was adjusted until the deflection became equal to the E_1 deflection, whereupon the value of E_1 was obtained from the value of the calibrating voltage as read on an alternating current voltmeter. As a further precaution, the calibration was made at the same place on the oscillograph screen that the value of E_1 was measured.

The experiments with the cathode-ray oscillograph indicate that for the copper arc in air the value of the initial voltage E_1 is about 12.2 volts. These measurements were made with an applied voltage of 120 volts d.c. and a load current of 5 amperes. An inductance of approximately 50 millihenries was connected in series with the circuit and shunted with a knife switch so that the inductance could be thrown into or out of the circuit. No change in the value of E_1 could be detected with or without the inductance.

Similar results were obtained when alternating current was used in place of direct current (see Fig. 3).

The initial voltage E_1 develops in less than 10^{-5} seconds. This was determined with the cathode-ray oscillograph with the beam moving horizontally at a rate of about 30,000 mm per second. If the trace of E_1 had been displaced 1 mm along the time axis, the development of E_1 would have occurred in 3.3×10^{-5} seconds. Actually, the voltage traces indicated at E_1 and, upon extinguishing the arc, at E_2 as shown in Fig. 4, appeared parallel to each other and showed no inclination toward the time axis. Measurements of the distance along the horizontal or time axis between E_1 and E_2 at the extremities of E_1 (Fig. 4) indicated close parallelism. From this it appears safe to conclude that E_1 develops in less than 10^{-5} seconds.

Discussion

The voltage drop across an electric arc is expended in maintaining the three major properties of the arc, the anode fall, the positive column drop, and the cathode fall. The anode fall and positive column drop depend upon either the arc current, the arc length or upon the size and shape of the arc terminals, and therefore, cannot be considered as arc characteristics unless the conditions are prescribed under which these variables assume definite values. On the other hand, the cathode fall is truly a characteristic property of the arc, inasmuch as its value is independent of these variables, and depends only upon the material of which the terminals are composed, and upon the gaseous atmosphere in which the arc is formed.

In the experiments described in this paper, it was found that the rapidly developed initial voltage E_1 is independent of arc current which suggested that this reproducible voltage is in reality a measure of the cathode fall of the arc. This raises the question as to why the anode fall should not contribute to the cathode fall in determining the initial arc voltage, E_1. In other words, is it proper to assume that the anode drop contributes negligibly to the initial voltage E_1 rather than that the initial voltage is the sum of the cathode and the anode falls? It appears reasonable to exclude the anode fall for the following reason: the usual concept of the anode fall of an arc is that, in the space adjacent to the anode, there is a more or less random distribution of ions and electrons, and that the variable nature of the anode fall, which in fact, may be either positive or negative, is due to the distribution of charges in this space. Should the requirements of the external circuit make necessary a larger current than would tend to flow through the arc for a particular distribution of ions and electrons, the anode fall would adjust itself to be positive in order to attract more electrons to the anode and thereby increase the current flow through the arc. If a smaller current than would tend to flow due to the distribution of ions and electrons, is needed to satisfy the requirements of the external circuit, a negative anode fall would arise in order to cause the actual current flow to meet the external circuit requirements. In the above experiments, in which the contacts carry current and then are separated, whereupon a voltage E_1 arises across the arc gap while the arc still has an infinitesimal length, the arc length at which the E_1 value is attained is believed to be less than the thickness of the cathode fall space. Therefore, the anode in this case is within the cathode fall space, and the random distribution of ions and electrons predicated in usual anode phenomena is not present and the anode fall does not exist.

Summary

1. Electromagnetic and cathode-ray oscillograph observations of the voltage arising across an arc gap when an arc is initiated indicate that there is a sudden rise of voltage after which there follows a more gradual change in arc voltage due to variations in arc length and arc current.

2. For the copper arc in air, the rapidly attained initial voltage, designated E_1, arises in less than 10^{-5} seconds as indicated by the cathode-ray oscillograph.

3. The initial voltage has a value of approximately 12.2 volts for copper contacts in air.

4. That the initial voltage E_1 may be associated with the cathode fall of the arc is suggested by the fact that the value of E_1 is independent of arc current throughout the current range investigated. The value of E_1 is not altered when the circuit voltage is 120 or 240 volts, either alternating or direct current.

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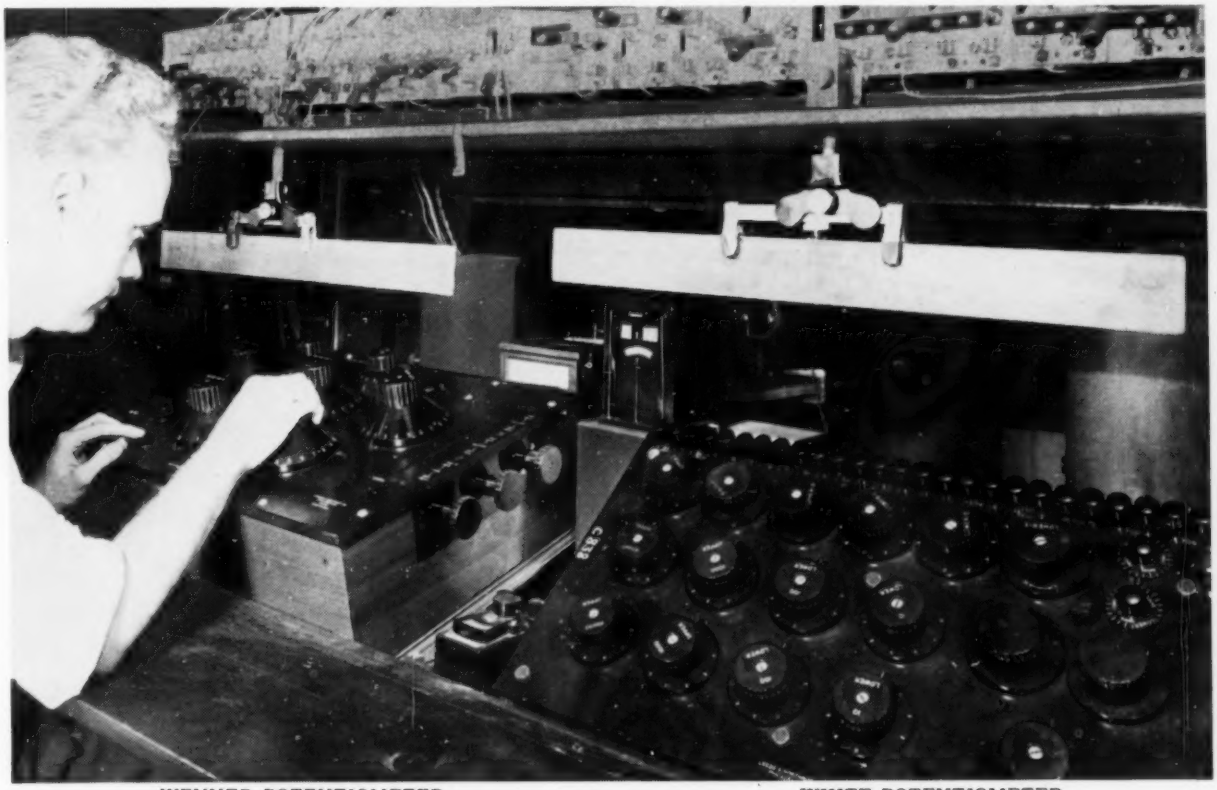

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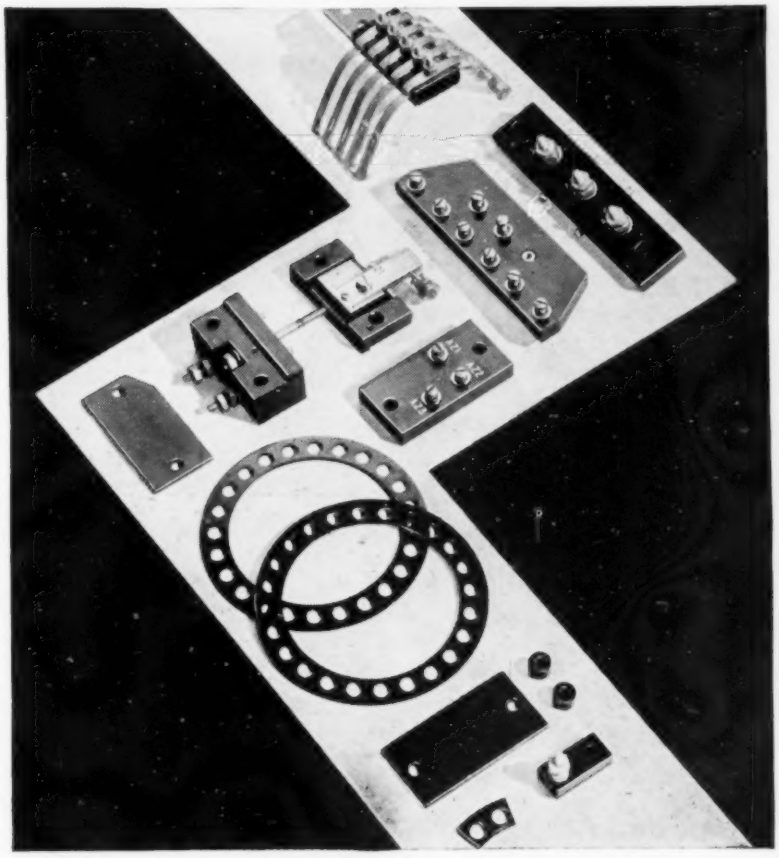

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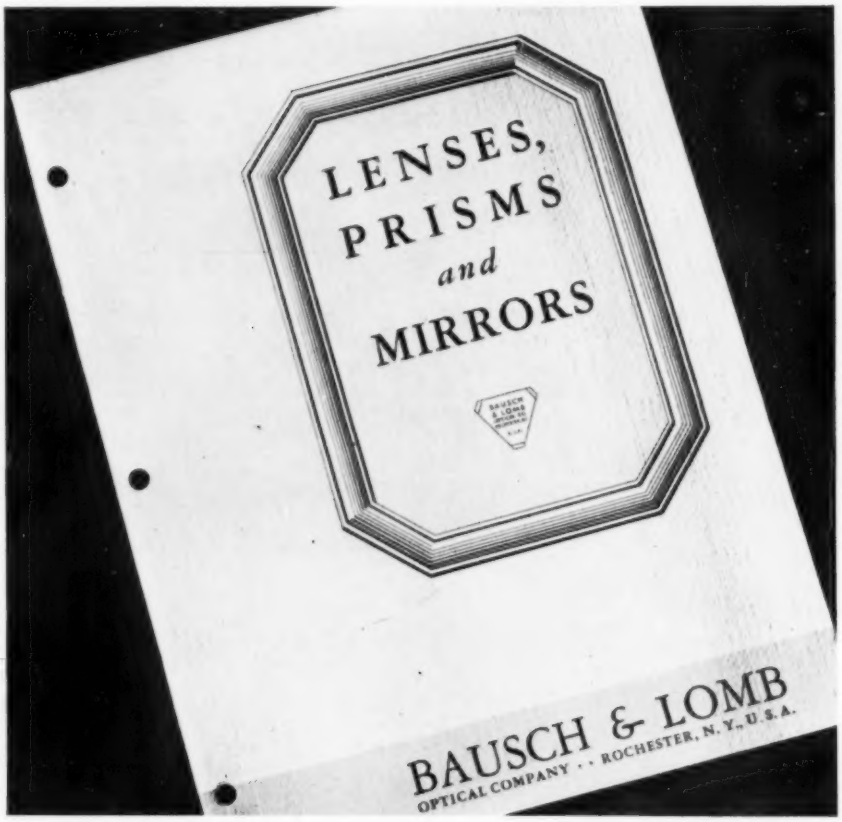

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THE Editors present for convenient reference in this section a list of those articles dealing with physics published in some thirty selected journals. Their definition of the scope of physics and their selection of the particular journals to be included are necessarily based on their interpretation of the wishes of the majority of the readers. The emphasis is placed on prompt mention and convenience of use rather than completeness. For many other excellent articles and publications, reference should be made to the regular abstract journals. Miss Margaret Shields, Physics Librarian at Princeton University, very kindly prepares the listings for the foreign language journals.

Annalen der Physik

SERIES 5, VOL. 30, NO. 5, SEPTEMBER 28, 1937.

Annihilation of Electron Space Charge by Positive Carriers. pp. 401–419. ***R. Kienzle***

Theory of Vibration of Crystal Plates. pp. 420–432. ***E. Lonn***

Electrical and Optical Behavior of Semi-Conductors: Cadmium, Thallium and Tin Oxides. pp. 433–445. ***G. Bauer***

Photo-Production of Electrons in Corona Discharge at Low Pressures. pp. 446–466. ***W. Christoph***

Magnetic Measurements on Cuprous Oxide. pp. 467–480. ***F. Hommel***

Astrophysical Journal

VOL. 86, NO. 4, NOVEMBER, 1937.

Sodium Content of the Head of the Great Daylight Comet Skjellerup 1927 K. pp. 345–362. ***Arthur Adel et al.***

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Photometry of the G Band in Representative Stellar Spectra. pp. 460–469. ***W. A. Rense and J. A. Hynek***

On the Origin of the Solar System. pp. 470–482. ***W. J. Luyten and E. L. Hill***

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VOL. 205, NO. 12, SEPTEMBER 20, 1937.

Magnetic Properties of Alloys of Iron and Iridium. pp. 517–518. ***M. Fallot***

Lα Satellites of Some Heavy Elements and Rare Earths. pp. 519–521. ***Y. Cauchois***

VOL. 205, NO. 14, OCTOBER 4, 1937.

Magnetic Properties of Alloys of Iron and Rhodium. pp. 558–560. ***M. Fallot***

Near Infrared Investigation of Ice and of Water and Its Solution in Dioxane. pp. 560–562. ***G. Bosschieter and J. Errera***

VOL. 205, NO. 15, OCTOBER 11, 1937.

Focusing of X-Rays by a Crystal of Variable Curvature. pp. 605–607. ***V. Dolejsek and M. Tayerle***

Proper Frequencies of Chlorates, Bromates and Iodates. pp. 607–609. ***M. Parodi***

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Continuous Recording of Cosmic Radiation on the Jungfraujoch. pp. 425–430. ***P. Scherrer et al.***

Journal de Physique

SERIES 7, VOL. 8, NO. 9, SEPTEMBER, 1937.

Electronic Analysis of the Oxidation of Gold-Copper Alloys. pp. 353–354. ***J. Trillat et al.***

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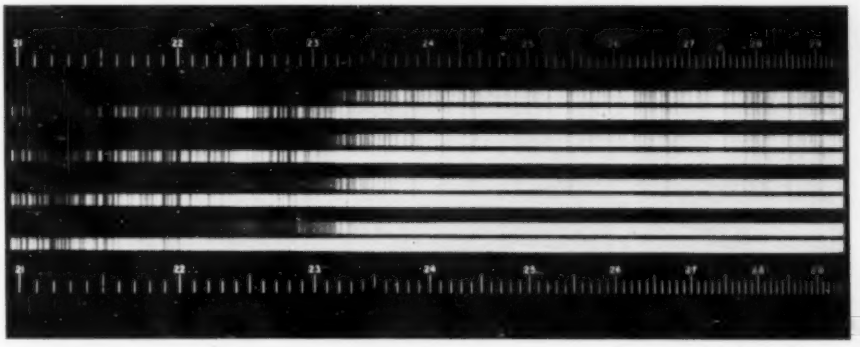

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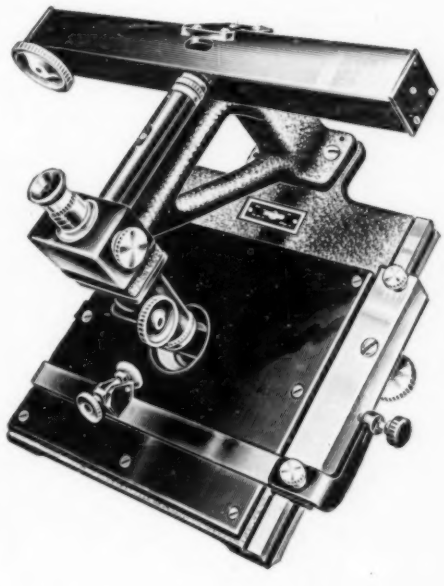

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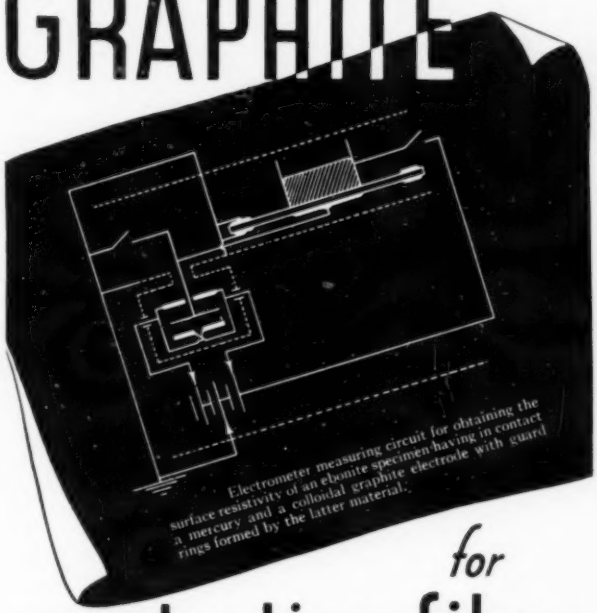

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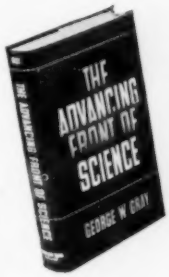

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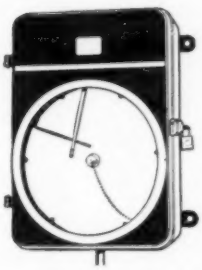

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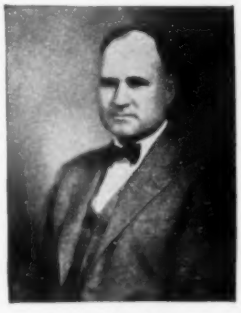

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